

PCT

8 Rec'd PCT/PTO REQUEST 15 DEC 1999

For receiving Office use only

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

International Application No. .

International Filing Date

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference  
(if desired) (12 characters maximum) N.74722 MAM

Box No. I TITLE OF INVENTION

PLASTIC COMPOSITIONS HAVING MINERAL-LIKE APPEARANCE

Box No. II APPLICANT

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)

ATO THAAS HOLDING C.V.  
SCHIPHOLPOORT 60, 2034 MB HAARLEM,  
THE NETHERLANDS

This person is also inventor.

Telephone No.

Facsimile No.

Teleprinter No.

State (i.e. country) of nationality:

NL

State (i.e. country) of residence:

NL

This person is applicant for the purposes of:

all designated States

all designated States except the United States of America

the United States of America only

the States indicated in the Supplemental Box

Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)

YANG, SHIJUN  
1803 PERRIN COURT  
MAPLE GLEN  
PENNYSYLVANIA 19002  
U.S.A.

This person is:

applicant only

applicant and inventor

inventor only (If this check-box is marked, do not fill in below.)

State (i.e. country) of nationality:

US

State (i.e. country) of residence:

US

This person is applicant for the purposes of:

all designated States

all designated States except the United States of America

the United States of America only

the States indicated in the Supplemental Box

Further applicants and/or (further) inventors are indicated on a continuation sheet.

Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:

agent

common representative

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

MARSHALL, MONICA ANNE,  
J.A. KEMP & CO.  
14 SOUTH SQUARE,  
GRAY'S INN, LONDON WC1R 5LX.  
UNITED KINGDOM.

Telephone No.

0171 405 3292

Facsimile No.

0171 242 8932

Teleprinter No.

Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

## Continuation of Box No. III FURTHER APPLICANTS AND/OR (FURTHER) INVENTORS

*If none of the following sub-boxes is used, this sheet is not to be included in the request.*

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)

KEATING, PAUL JOSEPH  
2727 AVENUE "A"  
NEWPORTVILLE  
PENNSYLVANIA 19056  
U.S.A.

This person is:

applicant only  
 applicant and inventor  
 inventor only (If this check-box is marked, do not fill in below.)

State (i.e. country) of nationality:

US

State (i.e. country) of residence:

US

This person is applicant for the purposes of:

 all designated States all designated States except the United States of America the United States of America only the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)

This person is:

applicant only  
 applicant and inventor  
 inventor only (If this check-box is marked, do not fill in below.)

State (i.e. country) of nationality:

State (i.e. country) of residence:

This person is applicant for the purposes of:

 all designated States all designated States except the United States of America the United States of America only the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)

This person is:

applicant only  
 applicant and inventor  
 inventor only (If this check-box is marked, do not fill in below.)

State (i.e. country) of nationality:

State (i.e. country) of residence:

This person is applicant for the purposes of:

 all designated States all designated States except the United States of America the United States of America only the States indicated in the Supplemental Box

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)

This person is:

applicant only  
 applicant and inventor  
 inventor only (If this check-box is marked, do not fill in below.)

State (i.e. country) of nationality:

State (i.e. country) of residence:

This person is applicant for the purposes of:

 all designated States all designated States except the United States of America the United States of America only the States indicated in the Supplemental Box Further applicants and/or (further) inventors are indicated on another continuation sheet.

## Box No.V DESIGNATION OF STATES

The following designations are hereby made under Rule 4.9(a) (mark the applicable check-boxes; at least one must be marked):

## Regional Patent

AP ARIPO Patent: GH Ghana, GM Gambia, KE Kenya, LS Lesotho, MW Malawi, SD Sudan, SZ Swaziland, UG Uganda, ZW Zimbabwe, and any other State which is a Contracting State of the Harare Protocol and of the PCT

EA Eurasian Patent: AM Armenia, AZ Azerbaijan, BY Belarus, KG Kyrgyzstan, KZ Kazakhstan, MD Republic of Moldova, RU Russian Federation, TJ Tajikistan, TM Turkmenistan, and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT

EP European Patent: AT Austria, BE Belgium, CH and LI Switzerland and Liechtenstein, DE Germany, DK Denmark, ES Spain, FI Finland, FR France, GB United Kingdom, GR Greece, IE Ireland, IT Italy, LU Luxembourg, MC Monaco, NL Netherlands, PT Portugal, SE Sweden, and any other State which is a Contracting State of the European Patent Convention and of the PCT

OA OAPI Patent: BF Burkina Faso, BJ Benin, CF Central African Republic, CG Congo, CI Côte d'Ivoire, CM Cameroon, GA Gabon, GN Guinea, ML Mali, MR Mauritania, NE Niger, SN Senegal, TD Chad, TG Togo, and any other State which is a member State of OAPI and a Contracting State of the PCT (if other kind of protection or treatment desired, specify on dotted line) . . . . .

## National Patent (if other kind of protection or treatment desired, specify on dotted line):

<input checked="" type="checkbox"/> AL Albania . . . . .	<input checked="" type="checkbox"/> LT Lithuania . . . . .
<input checked="" type="checkbox"/> AM Armenia . . . . .	<input checked="" type="checkbox"/> LU Luxembourg . . . . .
<input checked="" type="checkbox"/> AT Austria . . . . .	<input checked="" type="checkbox"/> LV Latvia . . . . .
<input checked="" type="checkbox"/> AU Australia . . . . .	<input checked="" type="checkbox"/> MD Republic of Moldova . . . . .
<input checked="" type="checkbox"/> AZ Azerbaijan . . . . .	<input checked="" type="checkbox"/> MG Madagascar . . . . .
<input checked="" type="checkbox"/> BA Bosnia and Herzegovina . . . . .	<input checked="" type="checkbox"/> MK The former Yugoslav Republic of Macedonia . . . . .
<input checked="" type="checkbox"/> BB Barbados . . . . .	<input checked="" type="checkbox"/> . . . . .
<input checked="" type="checkbox"/> BG Bulgaria . . . . .	<input checked="" type="checkbox"/> MN Mongolia . . . . .
<input checked="" type="checkbox"/> BR Brazil . . . . .	<input checked="" type="checkbox"/> MW Malawi . . . . .
<input checked="" type="checkbox"/> BY Belarus . . . . .	<input checked="" type="checkbox"/> MX Mexico . . . . .
<input checked="" type="checkbox"/> CA Canada . . . . .	<input checked="" type="checkbox"/> NO Norway . . . . .
<input checked="" type="checkbox"/> CH and LI Switzerland and Liechtenstein . . . . .	<input checked="" type="checkbox"/> NZ New Zealand . . . . .
<input checked="" type="checkbox"/> CN China . . . . .	<input checked="" type="checkbox"/> PL Poland . . . . .
<input checked="" type="checkbox"/> CU Cuba . . . . .	<input checked="" type="checkbox"/> PT Portugal . . . . .
<input checked="" type="checkbox"/> CZ Czech Republic . . . . .	<input checked="" type="checkbox"/> RO Romania . . . . .
<input checked="" type="checkbox"/> DE Germany . . . . .	<input checked="" type="checkbox"/> RU Russian Federation . . . . .
<input checked="" type="checkbox"/> DK Denmark . . . . .	<input checked="" type="checkbox"/> SD Sudan . . . . .
<input checked="" type="checkbox"/> EE Estonia . . . . .	<input checked="" type="checkbox"/> SE Sweden . . . . .
<input checked="" type="checkbox"/> ES Spain . . . . .	<input checked="" type="checkbox"/> SG Singapore . . . . .
<input checked="" type="checkbox"/> FI Finland . . . . .	<input checked="" type="checkbox"/> SI Slovenia . . . . .
<input checked="" type="checkbox"/> GB United Kingdom . . . . .	<input checked="" type="checkbox"/> SK Slovakia . . . . .
<input checked="" type="checkbox"/> GE Georgia . . . . .	<input checked="" type="checkbox"/> SL Sierra Leone . . . . .
<input checked="" type="checkbox"/> GH Ghana . . . . .	<input checked="" type="checkbox"/> TJ Tajikistan . . . . .
<input checked="" type="checkbox"/> GM Gambia . . . . .	<input checked="" type="checkbox"/> TM Turkmenistan . . . . .
<input checked="" type="checkbox"/> GW Guinea-Bissau . . . . .	<input checked="" type="checkbox"/> TR Turkey . . . . .
<input checked="" type="checkbox"/> HU Hungary . . . . .	<input checked="" type="checkbox"/> TT Trinidad and Tobago . . . . .
<input checked="" type="checkbox"/> ID Indonesia . . . . .	<input checked="" type="checkbox"/> UA Ukraine . . . . .
<input checked="" type="checkbox"/> IL Israel . . . . .	<input checked="" type="checkbox"/> UG Uganda . . . . .
<input checked="" type="checkbox"/> IS Iceland . . . . .	<input checked="" type="checkbox"/> US United States of America . . . . .
<input checked="" type="checkbox"/> JP Japan . . . . .	<input checked="" type="checkbox"/> . . . . .
<input checked="" type="checkbox"/> KE Kenya . . . . .	<input checked="" type="checkbox"/> UZ Uzbekistan . . . . .
<input checked="" type="checkbox"/> KG Kyrgyzstan . . . . .	<input checked="" type="checkbox"/> VN Viet Nam . . . . .
<input checked="" type="checkbox"/> KP Democratic People's Republic of Korea . . . . .	<input checked="" type="checkbox"/> YU Yugoslavia . . . . .
<input checked="" type="checkbox"/> KR Republic of Korea . . . . .	<input checked="" type="checkbox"/> ZW Zimbabwe . . . . .
<input checked="" type="checkbox"/> KZ Kazakhstan . . . . .	
<input checked="" type="checkbox"/> LC Saint Lucia . . . . .	
<input checked="" type="checkbox"/> LK Sri Lanka . . . . .	
<input checked="" type="checkbox"/> LR Liberia . . . . .	
<input checked="" type="checkbox"/> LS Lesotho . . . . .	

Check-boxes reserved for designating States (for the purposes of a national patent) which have become party to the PCT after issuance of this sheet:

CY Cyprus . . . . .

. . . . .

. . . . .

In addition to the designations made above, the applicant also makes under Rule 4.9(b) all designations which would be permitted under the PCT except the designation(s) of . . . . .

The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit. (Confirmation of a designation consists of the filing of a notice specifying that designation and the payment of the designation and confirmation fees. Confirmation must reach the receiving Office within the 15-month time limit.)

Supplemental B x *If the Supplemental Box is not used, this sheet need not be included in the request.**Use this box in the following cases:***1. If, in any of the Boxes, the space is insufficient to furnish all the information:***in particular:*

- (i) *if more than two persons are involved as applicants and/or inventors and no "continuation sheet" is available:*
- (ii) *if, in Box No. II or in any of the sub-boxes of Box No. III, the indication "the States indicated in the Supplemental Box" is checked:*
- (iii) *if, in Box No. II or in any of the sub-boxes of Box No. III, the inventor or the inventor/applicant is not inventor for the purposes of all designated States or for the purposes of the United States of America:*
- (iv) *if, in addition to the agent(s) indicated in Box No. IV, there are further agents:*
- (v) *if, in Box No. V, the name of any State (or OAPI) is accompanied by the indication "patent of addition," or "certificate of addition," or if, in Box No. V, the name of the United States of America is accompanied by an indication "Continuation" or "Continuation-in-part":*
- (vi) *if there are more than three earlier applications whose priority is claimed:*

**2. If the applicant claims, in respect of any designated Office, the benefits of provisions of the national law concerning non-prejudicial disclosures or exceptions to lack of novelty:****CONTINUATION OF BOX IV**

CANNON, David Lovell; ALLEN, William Guy Fairfax; GOLDIN, Douglas Michael; ELLIS-JONES, Patrick George Armine; BARLOW, Roy James; SENIOR, Alan Murray; BENTHAM, Stephen; AYERS, Martyn Lewis Stanley; WOODS, Geoffrey Corlett; CRESSWELL Thomas Anthony; MARSHALL, Monica Anne; WEBB, Andrew John; SEXTON, Jane Helen; NICHOLLS, Michael John; KEEN, Celia Mary; PRICE, Nigel John King; LEEMING, John Gerard; DUCKWORTH, Timothy John; IRVINE, Jonquil Claire; SMITH, Samuel Leonard; MCCLUSKIE, Gail Wilson; CURWEN, Julian Charles Barton; WRIGHT, Simon Mark; BENSON, John Everett; CLEEVE, James Harold Findlay; CAMPBELL, Patrick John; MERRYWEATHER, Colin Henry and DUCKETT, Anthony John; MIDGLEY, Jonathan Lee; BENTHAM, Andrew; and ROQUES; Sarah Elizabeth: of J.A. KEMP & CO., 14 South Square, Gray's Inn, London, WC1R 5LX, United Kingdom.

*in such case, write "Continuation of Box No. ...." [indicate the number of the Box] and furnish the information in the same manner as required according to the captions of the Box in which the space was insufficient;*

*in such case, write "Continuation of Box No. III" and indicate for each additional person the same type of information as required in Box No. III. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below;*

*in such case, write "Continuation of Box No. II" or "Continuation of Box No. III" or "Continuation of Boxes No. II and No. III" (as the case may be), indicate the name of the applicant(s) involved and, next to (each) such name, the State(s) (and/or, where applicable, ARIPO, Eurasian, European or OAPI patent) for the purposes of which the named person is applicant;*

*in such case, write "Continuation of Box No. II" or "Continuation of Box No. III" or "Continuation of Boxes No. II and No. III" (as the case may be), indicate the name of the inventor(s) and, next to (each) such name, the State(s) (and/or, where applicable, ARIPO, Eurasian, European or OAPI patent) for the purposes of which the named person is inventor;*

*in such case, write "Continuation of Box No. IV" and indicate for each further agent the same type of information as required in Box No. IV;*

*in such case, write "Continuation of Box No. V" and the name of each State involved (or OAPI), and after the name of each such State (or OAPI), the number of the parent title or parent application and the date of grant of the parent title or filing of the parent application;*

*in such case, write "Continuation of Box No. VI" and indicate for each additional earlier application the same type of information as required in Box No. VI.*

*in such case, write "Statement Concerning Non-Prejudicial Disclosures or Exceptions to Lack of Novelty" and furnish that statement below.*

**Box No. VI PRIORITY CLAIM**Further priority claims are indicated in the Supplemental Box 

The priority of the following earlier application(s) is hereby claimed:

Country (in which, or for which, the application was filed)	Filing Date (day/month/year)	Application No.	Office of filing (only for regional or international application)
item (1) US	19 06 97	60/050,213	
item (2)			
item (3)			

Mark the following check-box if the certified copy of the earlier application is to be issued by the Office which for the purposes of the present international application is the receiving Office (a fee may be required):

The receiving Office is hereby requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) identified above as item(s): \_\_\_\_\_

**Box No. VII INTERNATIONAL SEARCHING AUTHORITY**

Choice of International Searching Authority (ISA) (If two or more International Searching Authorities are competent to carry out the international search, indicate the Authority chosen; the two-letter code may be used): ISA / \_\_\_\_\_

Earlier search Fill in where a search (international, international-type or other) by the International Searching Authority has already been carried out or requested and the Authority is now requested to base the international search, to the extent possible, on the results of that earlier search. Identify such search or request either by reference to the relevant application (or the translation thereof) or by reference to the search request:

Country (or regional Office): Date (day/month/year): Number:

**Box No. VIII CHECK LIST**

This international application contains the following number of sheets:	This international application is accompanied by the item(s) marked below:	
1. request : 5 sheets	1. <input type="checkbox"/> separate signed power of attorney	5. <input checked="" type="checkbox"/> fee calculation sheet
2. description : 28 sheets	2. <input type="checkbox"/> copy of general power of attorney	6. <input type="checkbox"/> separate indications concerning deposited microorganisms
3. claims : 5 sheets	3. <input type="checkbox"/> statement explaining lack of signature	7. <input type="checkbox"/> nucleotide and/or amino acid sequence listing (diskette)
4. abstract : 1 sheets	4. <input checked="" type="checkbox"/> priority document(s) identified in Box No. VI as item(s):	8. <input type="checkbox"/> other (specify): _____
5. drawings : _____		
Total : 39 sheets		

Figure No. \_\_\_\_\_ of the drawings (if any) should accompany the abstract when it is published.

**Box No. IX SIGNATURE OF APPLICANT OR AGENT**

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request).

MARSHALL, MONICA ANNE  
AUTHORISED REPRESENTATIVE

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1. Date of actual receipt of the purported international application:	2. Drawings:
3. Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application:	<input type="checkbox"/> received: <input type="checkbox"/> not received:
4. Date of timely receipt of the required corrections under PCT Article 11(2):	
5. International Searching Authority specified by the applicant: ISA /	6. <input type="checkbox"/> Transmittal of search copy delayed until search fee is paid

For International Bureau use only

Date of receipt of the record copy  
by the International Bureau:

# PCT

## FEE CALCULATION SHEET Annex to the Request

For receiving Office use only

Applicant's or agent's  
file reference N.74722 MAM

International application No.

Date stamp of the receiving Office

Applicant  
ATOHAAS HOLDING C.V.

### CALCULATION OF PRESCRIBED FEES

1. TRANSMITTAL FEE . . . . . 200 DEM T

2. SEARCH FEE . . . . . 2200 DEM S

International search to be carried out by \_\_\_\_\_  
(If two or more International Searching Authorities are competent in relation to the international application, indicate the name of the Authority which is chosen to carry out the international search.)

### 3. INTERNATIONAL FEE

#### Basic Fee

The international application contains 39 sheets.

first 30 sheets . . . . . 800 DEM b<sub>1</sub>  
$$\frac{9}{\text{remaining sheets}} \times \frac{19}{\text{additional amount}} = 171 \text{ DEM} \quad \boxed{b_2}$$

Add amounts entered at b<sub>1</sub> and b<sub>2</sub> and enter total at B . . . . . 971 DEM B

#### Designation Fees

The international application contains 73 designations.

11 x 184 DEM = 2024 DEM D  
number of designation fees      amount of designation fee  
payable (maximum 11)

Add amounts entered at B and D and enter total at I . . . . . 2995 DEM I  
(Applicants from certain States are entitled to a reduction of 75% of the international fee. Where the applicant is (or all applicants are) so entitled, the total to be entered at I is 25% of the sum of the amounts entered at B and D.)

### 4. FEE FOR PRIORITY DOCUMENT . . . . .

### 5. TOTAL FEES PAYABLE

Add amounts entered at T, S, I and P, and enter total in the TOTAL box . . . . .

5395 DEM

TOTAL

The designation fees are not paid at this time.

### MODE OF PAYMENT

authorization to charge  
deposit account (see below)  
 cheque  
 postal money order

bank draft  
 cash  
 revenue stamps

coupons  
 other (specify):

### DEPOSIT ACCOUNT AUTHORIZATION (this mode of payment may not be available at all receiving Offices)

The RO/ EP  is hereby authorized to charge the total fees indicated above to my deposit account.

is hereby authorized to charge any deficiency or credit any overpayment in the total fees indicated above to my deposit account.

is hereby authorized to charge the fee for preparation and transmittal of the priority document to the International Bureau of WIPO to my deposit account.

2805.0038

12 06 98

Deposit Account Number

Date (day/month/year)

Signature

## PATENT COOPERATION TREATY

PCT/EP/38/03583

PCT

From the INTERNATIONAL BUREAU

NOTIFICATION OF RECEIPT OF  
RECORD COPY28 Rec'd PCT/PTO 15 DEC 1999  
(PCT Rule 24.2(a))

To:

MARSHALL, Monica, Anne  
J.A. Kemp & Co.  
14 South Square  
Gray's Inn  
London WC1R 5LX  
ROYAUME-UNI

MR.....

REC'D - 4 SEP 1998

DIARIED.....

Date of mailing (day/month/year) 26 August 1998 (26.08.98)	IMPORTANT NOTIFICATION
Applicant's or agent's file reference N.74722 MAM	International application No. PCT/EP98/03583

The applicant is hereby notified that the International Bureau has received the record copy of the international application as detailed below.

Name(s) of the applicant(s) and State(s) for which they are applicants:

ELF ATOCHEM S.A. (for all designated States except US)  
YANG, Shijun et al (for US)

International filing date : 15 June 1998 (15.06.98)  
Priority date(s) claimed : 19 June 1997 (19.06.97)  
Date of receipt of the record copy by the International Bureau : 12 August 1998 (12.08.98)  
List of designated Offices :

AP :GH,GM,KE,LS,MW,SD,SZ,UG,ZW  
EA :AM,AZ,BY,KG,KZ,MD,RU,TJ,TM  
EP :AT,BE,CH,CY,DE,DK,ES,FI,FR,GB,GR,IE,IT,LU,MC,NL,PT,SE  
OA :BF,BJ,CF,CG,CI,CM,GA,GN,ML,MR,NE,SN,TD,TG  
National :AL,AM,AT,AU,AZ,BA,BB,BG,BR,BY,CA,CH,CN,CU,CZ,DE,DK,EE,ES,FI,GB,GE,GH,GM,  
GW,HU,IL,IS,JP,KE,KG,KP,KR,KZ,LC,LK,LR,LS,LT,LU,LV,MD,MG,MK,MN,MW,MX,NO,NZ,PL,  
PT,RO,RU,SD,SE,SG,SI,SK,SL,TJ,TM,TR,TT,UA,UG,US,UZ,VN,YU,ZW

## ATTENTION

The applicant should carefully check the data appearing in this Notification. In case of any discrepancy between these data and the indications in the international application, the applicant should immediately inform the International Bureau.

In addition, the applicant's attention is drawn to the information contained in the Annex, relating to:

time limits for entry into the national phase  
 confirmation of precautionary designations  
 requirements regarding priority documents

A copy of this Notification is being sent to the receiving Office and to the International Searching Authority.

The International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland

Facsimile N. (41-22) 740.14.35

Authorized officer:

Aino Metcalfe

Telephone No. (41-22) 338.83.38

**INFORMATION ON TIME LIMITS FOR ENTERING THE NATIONAL PHASE**

The applicant is reminded that the "national phase" must be entered before each of the designated Offices indicated in the Notification of Receipt of Record Copy (Form PCT/IB/301) by paying national fees and furnishing translations, as prescribed by the applicable national laws.

The time limit for performing these procedural acts is **20 MONTHS** from the priority date or, for those designated States which the applicant elects in a demand for international preliminary examination or in a later election, **30 MONTHS** from the priority date, provided that the election is made before the expiration of 19 months from the priority date. Some designated (or elected) Offices have fixed time limits which expire even later than 20 or 30 months from the priority date. In other Offices an extension of time or grace period, in some cases upon payment of an additional fee, is available.

In addition to these procedural acts, the applicant may also have to comply with other special requirements applicable in certain Offices. It is the applicant's responsibility to ensure that the necessary steps to enter the national phase are taken in a timely fashion. Most designated Offices do not issue reminders to applicants in connection with the entry into the national phase.

For detailed information about the procedural acts to be performed to enter the national phase before each designated Office, the applicable time limits and possible extensions of time or grace periods, and any other requirements, see the relevant Chapters of Volume II of the PCT Applicant's Guide. Information about the requirements for filing a demand for international preliminary examination is set out in Chapter IX of Volume I of the PCT Applicant's Guide.

GR and ES became bound by PCT Chapter II on 7 September 1996 and 6 September 1997, respectively, and may, therefore, be elected in a demand or a later election filed on or after 7 September 1996 and 6 September 1997, respectively, regardless of the filing date of the international application. (See second paragraph above.)

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

**CONFIRMATION OF PRECAUTIONARY DESIGNATIONS**

This notification lists only specific designations made under Rule 4.9(a) in the request. It is important to check that these designations are correct. Errors in designations can be corrected where precautionary designations have been made under Rule 4.9(b). The applicant is hereby reminded that any precautionary designations may be confirmed according to Rule 4.9(c) before the expiration of 15 months from the priority date. If it is not confirmed, it will automatically be regarded as withdrawn by the applicant. There will be no reminder and no invitation. Confirmation of a designation consists of the filing of a notice specifying the designated State concerned (with an indication of the kind of protection or treatment desired) and the payment of the designation and confirmation fees. Confirmation must reach the receiving Office within the 15-month time limit.

**REQUIREMENTS REGARDING PRIORITY DOCUMENTS**

For applicants who have not yet complied with the requirements regarding priority documents, the following is recalled.

Where the priority of an earlier national, regional or international application is claimed, the applicant must submit a copy of the said earlier application, certified by the authority with which it was filed ("the priority document") to the receiving Office (which will transmit it to the International Bureau) or directly to the International Bureau, before the expiration of 16 months from the priority date, provided that any such priority document may still be submitted to the International Bureau before that date of international publication of the international application, in which case that document will be considered to have been received by the International Bureau on the last day of the 16-month time limit (Rule 17.1(a)).

Where the priority document is issued by the receiving Office, the applicant may, instead of submitting the priority document, request the receiving Office to prepare and transmit the priority document to the International Bureau. Such request must be made before the expiration of the 16-month time limit and may be subjected by the receiving Office to the payment of a fee (Rule 17.1(b)).

If the priority document concerned is not submitted to the International Bureau or if the request to the receiving Office to prepare and transmit the priority document has not been made (and the corresponding fee, if any, paid) within the applicable time limit indicated under the preceding paragraphs, any designated State may disregard the priority claim, provided that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity to furnish the priority document within a time limit which is reasonable under the circumstances.

Where several priorities are claimed, the priority date to be considered for the purposes of computing the 16-month time limit is the filing date of the earliest application whose priority is claimed.

# PATENT COOPERATION TREATY

From the INTERNATIONAL BUREAU

**PCT**

**NOTIFICATION CONCERNING  
SUBMISSION OR TRANSMITTAL  
OF PRIORITY DOCUMENT**

(PCT Administrative Instructions, Section 411)

Date of mailing (day/month/year)

26 August 1998 (26.08.98)

To:

MARSHALL, Monica, Anne  
J.A. Kemp & Co.  
14 South Square  
Gray's Inn  
London WC1R 5LX  
ROYAUME-UNI

Applicant's or agent's file reference

N.74722 MAM

**IMPORTANT NOTIFICATION**

International application No.

PCT/EP98/03583

International filing date (day/month/year)

15 June 1998 (15.06.98)

International publication date (day/month/year)

Not yet published

Priority date (day/month/year)

19 June 1997 (19.06.97)

Applicant

ELF ATOCHEM S.A. et al

1. The applicant is hereby notified of the date of receipt (except where the letters "NR" appear in the right-hand column) by the International Bureau of the priority document(s) relating to the earlier application(s) indicated below. Unless otherwise indicated by an asterisk appearing next to a date of receipt, or by the letters "NR", in the right-hand column, the priority document concerned was submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b).
2. This updates and replaces any previously issued notification concerning submission or transmittal of priority documents.
3. An asterisk(\*) appearing next to a date of receipt, in the right-hand column, denotes a priority document submitted or transmitted to the International Bureau but not in compliance with Rule 17.1(a) or (b). In such a case, the attention of the applicant is directed to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.
4. The letters "NR" appearing in the right-hand column denote a priority document which was not received by the International Bureau or which the applicant did not request the receiving Office to prepare and transmit to the International Bureau, as provided by Rule 17.1(a) or (b), respectively. In such a case, the attention of the applicant is directed to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.

<u>Priority date</u>	<u>Priority application No.</u>	<u>Country or regional Office or PCT receiving Office</u>	<u>Date of receipt of priority document</u>
19 June 1997 (19.06.97)	60/050,213	US	12 Augu 1998 (12.08.98)

The International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer

Aino Metcalfe

Telephone No. (41-22) 338.83.38

15 DEC 1999 PCT

28 Rec'd PCT/PTO

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>N.74722 MAM</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/EP 98/03583</b>	International filing date (day/month/year) <b>15/06/1998</b>	(Earliest) Priority Date (day/month/year) <b>19/06/1997</b>
Applicant <b>ELF ATOCHEM S.A. et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of **02** sheets.

It is also accompanied by a copy of each prior art document cited in this report.

1.  Certain claims were found unsearchable (see Box I).
2.  Unity of invention is lacking (see Box II).
3.  The international application contains disclosure of a nucleotide and/or amino acid sequence listing and the international search was carried out on the basis of the sequence listing
  - filed with the international application.
  - furnished by the applicant separately from the international application,
    - but not accompanied by a statement to the effect that it did not include matter going beyond the disclosure in the international application as filed.
  - Transcribed by this Authority
4. With regard to the title,
  - the text is approved as submitted by the applicant
  - the text has been established by this Authority to read as follows:  
**plastic composition**
5. With regard to the abstract,
  - the text is approved as submitted by the applicant
  - the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this International Search Report, submit comments to this Authority.
6. The figure of the drawings to be published with the abstract is:
 

Figure No. ---

  - as suggested by the applicant.
  - because the applicant failed to suggest a figure.
  - because this figure better characterizes the invention.

None of the figures.

A. CL/ **IFICATION OF SUBJECT MATTER**  
 IPC C C08L101/00 B29C47/10 // (C08L101/00, 101:00)

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08L B29C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 97 14749 A (ICI ACRYLICS INC) 24 April 1997 cited in the application see page 9, line 5 - line 25 -----	1



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

\* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention
- "X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone
- "Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.
- "&" document member of the same patent family

Date of the actual completion of the international search

26 October 1998

Date of mailing of the international search report

16/11/1998

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl.  
Fax: (+31-70) 340-3016

Authorized officer

Schueler, D

**INTERNATIONAL SEARCH REPORT**

Information on patent family members

International Application No

PCT/EP 98/03583

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
WO 9714749	A 24-04-1997	AU 7432696	A 07-05-1997	EP 0856032 A 05-08-1998

PATENT COOPERATION TREATY

WO 98/59006  
PCT/EP98/03583

8 Rec'd PCT PCTO 15 DEC 1999

From the INTERNATIONAL BUREAU

To:

MARSHALL, Monica, Anne  
J.A. Kemp & Co.  
14 South Square  
Gray's Inn  
London WC1R 5LX  
ROYAUME-UNI

*TO MAM*

NOTICE INFORMING THE APPLICANT OF THE  
COMMUNICATION OF THE INTERNATIONAL  
APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

Date of mailing (day/month/year)

30 December 1998 (30.12.98)

Applicant's or agent's file reference

N.74722 MAM

IMPORTANT NOTICE

International application No.

PCT/EP98/03583

International filing date (day/month/year)

15 June 1998 (15.06.98)

Priority date (day/month/year)

19 June 1997 (19.06.97)

Applicant

ELF ATOCHEM S.A. et al

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:

AU, BR, CA, CN, EP, IL, JP, KP, KR, PL, US

In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:

AL, AM, AP, AT, AZ, BA, BB, BG, BY, CH, CU, CZ, DE, DK, EA, EE, ES, FI, GB, GE, GH, GM, GW, HU, ID, IS, KE, KG, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, OA, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, UZ, VN, YU, ZW

The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on 30 December 1998 (30.12.98) under No. WO 98/59006

**REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)**

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a demand for international preliminary examination must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

**REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))**

If the applicant wishes to proceed with the international application in the national phase, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO  
34, chemin des Colombettes  
1211 Genève 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer

J. Zahra

Telephone No. (41-22) 338.83.38

## Continuation of Form PCT/IB/308

NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF  
THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES

Date of mailing (day/month/year) 30 December 1998 (30.12.98)	<b>IMPORTANT NOTICE</b>
Applicant's or agent's file reference N.74722 MAM	International application No. PCT/EP98/03583
<p>The applicant is hereby notified that, at the time of establishment of this Notice, the time limit under Rule 46.1 for making amendments under Article 19 has not yet expired and the International Bureau had received neither such amendments nor a declaration that the applicant does not wish to make amendments.</p>	

The demand must be filed directly with the competent International Preliminary Examining Authority or, if two or more Authorities are competent, with the one chosen by the applicant. The full name or two-letter code of that Authority may be indicated by the applicant on the line below:  
IPF \*\* EP

8 Rec'd PCT/PTO 15 DEC 1999 PCT  
DEMAND

CHAPTER II

under Article 31 of the Patent Cooperation Treaty:

The undersigned requests that the international application specified below be the subject of international preliminary examination according to the Patent Cooperation Treaty and hereby elects all eligible States (except where otherwise indicated).

For International Preliminary Examining Authority use only

Identification of IPEA	Date of receipt of DEMAND
<b>Box No. I IDENTIFICATION OF THE INTERNATIONAL APPLICATION</b>	
International application No. PCT/EP98/03583	International filing date (day/month/year) 15 June 1998
(Earliest) Priority date (day/month/year) 19 June 1997	
Title of invention PLASTIC COMPOSITION	
<b>Box No. II APPLICANT(S)</b>	
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)  ELF ATOCHEM S.A. 4 & 8 Cours Michelet La Defense 10 F-92080 Puteaux France	Telephone No.:  Facsimile No.:  Teleprinter No.:
State (that is, country) of nationality: FR	State (that is, country) of residence: FR
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)  YANG, SHIJUN 1803 Perrin Court Maple Glen Pennsylvania 19002 US	
State (that is, country) of nationality: CN	State (that is, country) of residence: US
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)  KEATING, PAUL JOSEPH 2727 Avenue "A" Newportville Pennsylvania 19056 US	
State (that is, country) of nationality: US	State (that is, country) of residence: US
<input type="checkbox"/> Further applicants are indicated on a continuation sheet.	

## Box N . III AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The following person is  agent  common representative

and  has been appointed earlier and represents the applicant(s) also for international preliminary examination.

is hereby appointed and any earlier appointment of (an) agent(s)/common representative is hereby revoked.

is hereby appointed, specifically for the procedure before the International Preliminary Examining Authority, in addition to the agent(s)/common representative appointed earlier.

Name and address: *(Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)*

MARSHALL, Monica Anne  
J.A. KEMP & CO.,  
14 South Square,  
Gray's Inn,  
London, WC1R 5LX,  
United Kingdom

Telephone No.:

+44 171 405 3292

Facsimile No.:

+44 171 242 8932

Teleprinter No.:

23676

Address for correspondence: Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

## Box No. IV BASIS FOR INTERNATIONAL PRELIMINARY EXAMINATION

## Statement concerning amendments:\*

1. The applicant wishes the international preliminary examination to start on the basis of:

the international application as originally filed

the description  as originally filed

as amended under Article 34

the claims  as originally filed

as amended under Article 19 (together with any accompanying statement)

as amended under Article 34

the drawings  as originally filed

as amended under Article 34

2.  The applicant wishes any amendment to the claims under Article 19 to be considered as reversed.

3.  The applicant wishes the start of the international preliminary examination to be postponed until the expiration of 20 months from the priority date unless the International Preliminary Examining Authority receives a copy of any amendments made under Article 19 or a notice from the applicant that he does not wish to make such amendments (Rule 69.1(d)). *(This check-box may be marked only where the time limit under Article 19 has not yet expired.)*

\* Where no check-box is marked, international preliminary examination will start on the basis of the international application as originally filed or, where a copy of amendments to the claims under Article 19 and/or amendments of the international application under Article 34 are received by the International Preliminary Examining Authority before it has begun to draw up a written opinion or the international preliminary examination report, as so amended.

Language for the purposes of international preliminary examination: English

which is the language in which the international application was filed.

which is the language of a translation furnished for the purposes of international search.

which is the language of publication of the international application.

which is the language of the translation (to be) furnished for the purposes of international preliminary examination.

## Box No. V ELECTION OF STATES

The applicant hereby elects all eligible States *(that is, all States which have been designated and which are bound by Chapter II of the PCT)*

excluding the following States which the applicant wishes not to elect:

## Box No. VI CHECK LIST

The demand is accompanied by the following elements, in the language referred to in Box No. IV, for the purposes of international preliminary examination:

1. translation of international application	:	sheets
2. amendments under Article 34	:	sheets
3. copy (or, where required, translation) of amendments under Article 19	:	sheets
4. copy (or, where required, translation) of statement under Article 19	:	sheets
5. letter	:	sheets
6. other (specify)	:	sheets

For International Preliminary Examining Authority use only

received	not received
----------	--------------

<input type="checkbox"/>	<input type="checkbox"/>

The demand is also accompanied by the item(s) marked below:

1. <input checked="" type="checkbox"/> fee calculation sheet	4. <input type="checkbox"/> statement explaining lack of signature
2. <input type="checkbox"/> separate signed power of attorney	5. <input type="checkbox"/> nucleotide and or amino acid sequence listing in computer readable form
3. <input type="checkbox"/> copy of general power of attorney; reference number, if any:	6. <input type="checkbox"/> other (specify):

## Box No. VII SIGNATURE OF APPLICANT, AGENT OR COMMON REPRESENTATIVE

Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the demand).

MARSHALL, Monica Anne

## For International Preliminary Examining Authority use only

1. Date of actual receipt of DEMAND:

2. Adjusted date of receipt of demand due to CORRECTIONS under Rule 60.1(b):

3.  The date of receipt of the demand is AFTER the expiration of 19 months from the priority date and item 4 or 5, below, does not apply.

The applicant has been informed accordingly.

4.  The date of receipt of the demand is WITHIN the period of 19 months from the priority date as extended by virtue of Rule 80.5.

5.  Although the date of receipt of the demand is after the expiration of 19 months from the priority date, the delay in arrival is EXCUSED pursuant to Rule 82.

## For International Bureau use only

Demand received from IPEA on:

## PCT

## FEE CALCULATION SHEET

## Annex to the Demand for international preliminary examination

International application No. PCT/EP98/03583	For International Preliminary Examining Authority use only																		
Applicant's or agent's file reference N.74722 MAM/lp	Date stamp of the IPEA																		
Applicant ELF ATOCHEM S.A. et al.																			
<p><b>Calculation of prescribed fees</b></p> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 60%; padding: 5px;">1. Preliminary examination fee .....</td> <td style="width: 20%; text-align: center; padding: 5px;">DEM 3000</td> <td style="width: 20%; text-align: center; padding: 5px;">P</td> </tr> <tr> <td colspan="3" style="padding: 10px;"> <p>2. Handling fee (<i>Applicants from certain States are entitled to a reduction of 75% of the handling fee. Where the applicant is (or all applicants are) so entitled, the amount to be entered at H is 25% of the handling fee.</i>) .....</p> </td> </tr> <tr> <td colspan="3" style="text-align: center; padding: 5px;">DEM 285 H</td> </tr> <tr> <td colspan="3" style="padding: 10px;"> <p>3. Total of prescribed fees Add the amounts entered at P and H and enter total in the TOTAL box .....</p> </td> </tr> <tr> <td colspan="3" style="text-align: center; padding: 5px;">DEM 3285</td> </tr> <tr> <td colspan="3" style="text-align: center; padding: 5px;">TOTAL</td> </tr> </table>		1. Preliminary examination fee .....	DEM 3000	P	<p>2. Handling fee (<i>Applicants from certain States are entitled to a reduction of 75% of the handling fee. Where the applicant is (or all applicants are) so entitled, the amount to be entered at H is 25% of the handling fee.</i>) .....</p>			DEM 285 H			<p>3. Total of prescribed fees Add the amounts entered at P and H and enter total in the TOTAL box .....</p>			DEM 3285			TOTAL		
1. Preliminary examination fee .....	DEM 3000	P																	
<p>2. Handling fee (<i>Applicants from certain States are entitled to a reduction of 75% of the handling fee. Where the applicant is (or all applicants are) so entitled, the amount to be entered at H is 25% of the handling fee.</i>) .....</p>																			
DEM 285 H																			
<p>3. Total of prescribed fees Add the amounts entered at P and H and enter total in the TOTAL box .....</p>																			
DEM 3285																			
TOTAL																			
<p><b>Mode of Payment</b></p> <table style="width: 100%; border-collapse: collapse;"> <tr> <td style="width: 50%; padding: 5px;"> <input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)           </td> <td style="width: 50%; text-align: center; padding: 5px;"> <input type="checkbox"/> cash           </td> </tr> <tr> <td style="padding: 5px;"> <input type="checkbox"/> cheque           </td> <td style="text-align: center; padding: 5px;"> <input type="checkbox"/> revenue stamps           </td> </tr> <tr> <td style="padding: 5px;"> <input type="checkbox"/> postal money order           </td> <td style="text-align: center; padding: 5px;"> <input type="checkbox"/> coupons           </td> </tr> <tr> <td style="padding: 5px;"> <input type="checkbox"/> bank draft           </td> <td style="text-align: center; padding: 5px;"> <input type="checkbox"/> other (<i>specify</i>): _____           </td> </tr> </table>		<input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)	<input type="checkbox"/> cash	<input type="checkbox"/> cheque	<input type="checkbox"/> revenue stamps	<input type="checkbox"/> postal money order	<input type="checkbox"/> coupons	<input type="checkbox"/> bank draft	<input type="checkbox"/> other ( <i>specify</i> ): _____										
<input checked="" type="checkbox"/> authorization to charge deposit account with the IPEA (see below)	<input type="checkbox"/> cash																		
<input type="checkbox"/> cheque	<input type="checkbox"/> revenue stamps																		
<input type="checkbox"/> postal money order	<input type="checkbox"/> coupons																		
<input type="checkbox"/> bank draft	<input type="checkbox"/> other ( <i>specify</i> ): _____																		
<p><b>Deposit Account Authorization</b> (<i>this mode of payment may not be available at all IPEAs</i>)</p> <p>The IPEA/ EP <input checked="" type="checkbox"/> is hereby authorized to charge the total fees indicated above to my deposit account.</p> <p><input type="checkbox"/> (<i>this check-box may be marked only if the conditions for deposit accounts of the IPEA so permit</i>) is hereby authorized to charge any deficiency or credit any overpayment in the total fees indicated above to my deposit account.</p>																			
2805.0038	13 January 1999																		
Deposit Account Number	Date (day/month/year)	Signature																	

## PATENT COOPERATION TREATY

From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

PCT

To:

MARSHALL, Monica Anne  
 J. A. KEMP & CO.  
 14 Southampton Square  
 Gray's Inn  
 London WC1R 5LX  
 GRANDE BRETAGNE

3 Post PCT/PTO 15 DEC 1999

NOTIFICATION OF RECEIPT  
OF DEMAND BY COMPETENT INTERNATIONAL  
PRELIMINARY EXAMINING AUTHORITY(PCT Rules 59.3(e) and 61.1(b), first sentence  
and Administrative Instructions, Section 601(a))

		Date of mailing (day/month/year)	02.02.99
Applicant's or agent's file reference N. 74722 MAM/1p		IMPORTANT NOTIFICATION	
International application No. PCT/EP 98/03583	International filing date (day/month/year) 15/06/1998	Priority date (day/month/year) 19/06/1997	
Applicant ELF ATOCHEM S.A. et al.			

1. The applicant is hereby notified that this International Preliminary Examining Authority considers the following date as the date of receipt of the demand for international preliminary examination of the international application:

15/01/1999

2. This date of receipt is:



the actual date of receipt of the demand by this Authority (Rule 61.1(b)).



the actual date of receipt of the demand on behalf of this Authority (Rule 59.3(e)).



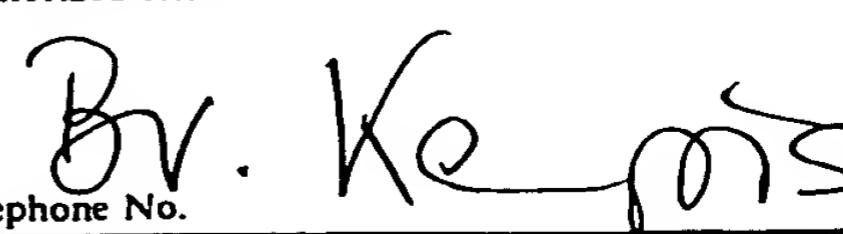
the date on which this Authority has, in response to the invitation to correct defects in the demand (Form PCT/IPEA/404), received the required corrections.

3.  ATTENTION: That date of receipt is AFTER the expiration of 19 months from the priority date. Consequently, the election(s) made in the demand does (do) not have the effect of postponing the entry into the national phase until 30 months from the priority date (or later in some Offices) (Article 39(1)). Therefore, the acts for entry into the national phase must be performed within 20 months from the priority date (or later in some Offices) (Article 22). For details, see the *PCT Applicant's Guide*, Volume II.



(If applicable) This notification confirms the information given by telephone, facsimile transmission or in person on:

4. Only where paragraph 3 applies, a copy of this notification has been sent to the International Bureau.

Name and mailing address of the IPEA/  European Patent Office D-80298 Munich Tel. (+49-89) 2399-0, Tx: 523656 epmu d Fax: (+49-89) 2399-4465	Authorized officer  Telephone No.
--	--

## PATENT COOPERATION TREATY

MAM

PCT

From the INTERNATIONAL BUREAU

INFORMATION CONCERNING ELECTED  
OFFICES NOTIFIED OF THEIR ELECTION

(PCT Rule 61.3)

To:

MARSHALL, Monica, Anne  
 J.A. Kemp & Co.  
 14 South Square  
 Gray's Inn  
 London WC1R 5LX  
 ROYAUME-UNI

Date of mailing (day/month/year)

05 February 1999 (05.02.99)

Applicant's or agent's file reference

N.74722 MAM

## IMPORTANT INFORMATION

International application No.

PCT/EP98/03583

International filing date (day/month/year)

15 June 1998 (15.06.98)

Priority date (day/month/year)

19 June 1997 (19.06.97)

Applicant

ELF ATOCHEM S.A. et al

1. The applicant is hereby informed that the International Bureau has, according to Article 31(7), notified each of the following Offices of its election:

AP :GH,GM,KE,LS,MW,SD,SZ,UG,ZW

EP :AT,BE,CH,CY,DE,DK,ES,FI,FR,GB,GR,IE,IT,LU,MC,NL,PT,SE

National :AU,BG,BR,CA,CN,CZ,DE,GB,IL,JP,KP,KR,MN,NO,NZ,PL,RO,RU,SE,SK,US,  
VN

2. The following Offices have waived the requirement for the notification of their election; the notification will be sent to them by the International Bureau only upon their request:

EA :AM,AZ,BY,KG,KZ,MD,RU,TJ,TM

OA :BF,BJ,CF,CG,CI,CM,GA,GN,ML,MR,NE,SN,TD,TG

National :AL,AM,AT,AZ,BA,BB,BY,CH,CU,DK,EE,ES,FI,GE,GH,GM,GW,HU,IS,KE,  
KG,KZ,LC,LK,LR,LS,LT,LU,LV,MD,MG,MK,MW,MX,PT,SD,SG,SI,SL,TJ,TM,TR,TT,UA,  
UG,UZ,YU,ZW

3. The applicant is reminded that he must enter the "national phase" before the expiration of 30 months from the priority date before each of the Offices listed above. This must be done by paying the national fee(s) and furnishing, if prescribed, a translation of the international application (Article 39(1)(a)), as well as, where applicable, by furnishing a translation of any annexes of the international preliminary examination report (Article 36(3)(b) and Rule 74.1).

Some offices have fixed time limits expiring later than the above-mentioned time limit. For detailed information about the applicable time limits and the acts to be performed upon entry into the national phase before a particular Office, see Volume II of the PCT Applicant's Guide.

The entry into the European regional phase is postponed until 31 months from the priority date for all States designated for the purposes of obtaining a European patent.

The International Bureau of WIPO  
 34, chemin des Colombettes  
 1211 Geneva 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer:

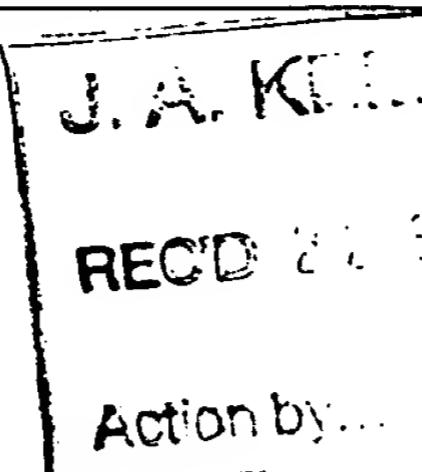
A. Karkachi

Telephone No. (41-22) 338.83.38

From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

MARSHALL, Monica Anne  
J.A. KEMP & CO.  
14 South Square  
Gray's Inn  
London WC1R 5LX  
GRANDE BRETAGNE



PCT

NOTIFICATION OF TRANSMITTAL OF  
THE INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT  
(PCT Rule 71.1)

Date of mailing  
(day/month/year)

20.09.99

Applicant's or agent's file reference  
N.74722 MAM/lp

IMPORTANT NOTIFICATION

International application No.  
PCT/EP98/03583

International filing date (day/month/year)  
15/06/1998

Priority date (day/month/year)  
19/06/1997

Applicant

ELF ATOCHEM S.A. et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.
4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/

European Patent Office  
D-80298 Munich  
Tel. +49 89 2399 - 0 Tx: 523656 epmu d  
Fax: +49 89 2399 - 4465

Authorized officer

Hardy Magliano N

Tel. +49 89 2399-8151



## PATENT COOPERATION TREATY

From the RECEIVING OFFICE

PCT

To:

Marshall, Monica Anne  
 J.A. KEMP & CO.  
 14 South Square  
 Gray's Inn  
 London WC1R 5EP  
 FRANCE BRETAGNE

28 Dec PCT TO 15 DEC 1999

NOTIFICATION OF DECISION CONCERNING  
REQUEST FOR RECTIFICATION

(PCT Rule 91.1(f))

		Date of mailing (day/month/year)	07.08.98
Applicant's or agent's file reference  N. 74722 MAM	REPLY DUE	NONE However, see last paragraph below	
International application No.  PCT/ EP 98/ 03583	International filing date (day/month/year)	15/06/1998	
Applicant  ELF ATOCHEM S.A.			

The applicant is hereby notified that this receiving Office has considered the request for rectification of obvious errors in the request of the international application and that it has decided :

1.  to authorize the rectification :  
 as requested by the applicant  
 to the extent set forth below\* :

2.  to refuse to authorize the rectification or part of it for the following reasons\* :

A copy of this notification, together with a copy of the applicant's request for rectification, has been sent to the International Bureau.

\* If the authorization of the rectification has been refused in whole or in part, the applicant may request the International Bureau, before the technical preparations for international publication have been completed and subject to the payment of a fee, to publish the request for rectification together with the international application. See Rule 91.1(f), third and fourth sentences, and, for the amount of the fee, see Annex B2(WO), Volume I of the PCT Applicant's Guide.

Name and mailing address of the receiving Office   European Patent Office, P.B. 5818 Patendaan 2 NL-2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 cpo nl. Fax: (+31-70) 340-3016	Authorized officer   P.L.R. Pether
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PCT

REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

For receiving Office use only

PCT/EP 98/03583

International Application No.

(15.06.98)

15 JUN 1998

International Filing Date

EUROPEAN PATENT OFFICE

PCT INTERNATIONAL APPLICATION

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference

(if desired) (12 characters maximum) N.74722 MAM

## Box No. I TITLE OF INVENTION

PLASTIC COMPOSITIONS HAVING MINERAL-LIKE APPEARANCE

## Box No. II APPLICANT

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)

ATOCHAAG HOLDING B.V.  
SCHIPHOLPOORT 60, 2034 MB HAARLEM,  
THE NETHERLANDS

ELF ATOCHEM S.A.  
4 & 8 Cours Michelet,  
La Defense 10,  
FR- 92080 Puteaux

 This person is also inventor.

Telephone No.

Facsimile No.

Teleprinter No.

/EP

/EP

State (i.e. country) of nationality:

[NL] FR

State (i.e. country) of residence:

[NL] FR

This person is applicant for the purposes of:

 all designated States all designated States except the United States of America the United States of America only the States indicated in the Supplemental Box

## Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)

YANG, SHIJUN  
1803 PERRIN COURT  
MAPLE GLEN  
PENNSYLVANIA 19002  
U.S.A.

This person is:

 applicant only applicant and inventor inventor only (If this check-box is marked, do not fill in below.)

/EP

State (i.e. country) of nationality:

[US] CA

State (i.e. country) of residence:

US

This person is applicant for the purposes of:

 all designated States all designated States except the United States of America the United States of America only the States indicated in the Supplemental Box

/EP

 Further applicants and/or (further) inventors are indicated on a continuation sheet.

## Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE

The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as:

 agent common representative

Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)

MARSHALL, MONICA ANNE,  
J.A. KEMP & CO.  
14 SOUTH SQUARE,  
GRAY'S INN, LONDON WC1R 5LX.  
UNITED KINGDOM.

Telephone No.

0171 405 3292

Facsimile No.

0171 242 8932

Teleprinter No.

 Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.

## PATENT COOPERATION TREATY

## PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>N.74722 MAM</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/EP 98/ 03583</b>	International filing date (day/month/year) <b>15/06/1998</b>	(Earliest) Priority Date (day/month/year) <b>19/06/1997</b>
Applicant <b>ELF ATOCHEM S.A. et al.</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 02 sheets.

It is also accompanied by a copy of each priorart document cited in this report.

1.  Certain claims were found unsearchable(see Box I).
2.  Unity of Invention is lacking(see Box II).
3.  The international application contains disclosure of a **nucleotide and/or amino acid sequence listing** and the international search was carried out on the basis of the sequence listing
  - filed with the international application.
  - furnished by the applicant separately from the international application,
    - but not accompanied by a statement to the effect that it did not include matter going beyond the disclosure in the international application as filed.
    - Transcribed by this Authority
4. With regard to the title,  the text is approved as submitted by the applicant  
 the text has been established by this Authority to read as follows:  
**plastic composition**
5. With regard to the abstract,
  - the text is approved as submitted by the applicant
  - the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this International Search Report, submit comments to this Authority.
6. The figure of the drawings to be published with the abstract is:
 

Figure No. ---

  - as suggested by the applicant.
  - because the applicant failed to suggest a figure.
  - because this figure better characterizes the invention.

None of the figures.

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C08L101/00 B29C47/10 // (C08L101/00, 101:00)

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08L B29C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 97 14749 A (ICI ACRYLICS INC) 24 April 1997 cited in the application see page 9, line 5 - line 25 -----	1

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

## \* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

26 October 1998

Date of mailing of the international search report

16/11/1998

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl.  
Fax: (+31-70) 340-3016

Authorized officer

Schueler, D

## INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/EP 98/03583

Patent document cited in search report	Publication date	Patent family member(s)		Publication date
WO 9714749	A 24-04-1997	AU 7432696	A 07-05-1997	EP 0856032 A 05-08-1998

## PATENT COOPERATION TREATY

PCT

From the INTERNATIONAL BUREAU

NOTIFICATION OF ELECTION  
(PCT Rule 61.2)

To:

United States Patent and Trademark  
Office  
(B x PCT)  
Crystal Plaza 2  
Washington, DC 20231  
ÉTATS-UNIS D'AMÉRIQUE

in its capacity as elected Office

Date of mailing (day/month/year) 05 February 1999 (05.02.99)	
International application No. PCT/EP98/03583	Applicant's or agent's file reference N.74722 MAM
International filing date (day/month/year) 15 June 1998 (15.06.98)	Priority date (day/month/year) 19 June 1997 (19.06.97)
Applicant YANG, Shijun et al	

## 1. The designated Office is hereby notified of its election made:

in the demand filed with the International Preliminary Examining Authority on:

15 January 1999 (15.01.99)

in a notice effecting later election filed with the International Bureau on:

\_\_\_\_\_

2. The election  was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland Facsimile No.: (41-22) 740.14.35	Authorized officer A. Karkachi Telephone No.: (41-22) 338.83.38
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## PATENT COOPERATION TREATY

## PCT

INTERNATIONAL PRELIMINARY EXAMINATION REPORT  
(PCT Article 36 and Rule 70)

Applicant's or agent's file reference  N.74722 MAM/Ip	<b>FOR FURTHER ACTION</b>		See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No.  PCT/EP98/03583	International filing date (day/month/year)  15/06/1998	Priority date (day/month/year)  19/06/1997	
International Patent Classification (IPC) or national classification and IPC  C08L101/00			
Applicant  ELF ATOCHEM S.A. et al.			

<p>1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.</p> <p>2. This REPORT consists of a total of 6 sheets, including this cover sheet.</p> <p><input type="checkbox"/> This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).</p> <p>These annexes consist of a total of sheets.</p>
<p>3. This report contains indications relating to the following items:</p> <ul style="list-style-type: none"> <li>I <input checked="" type="checkbox"/> Basis of the report</li> <li>II <input type="checkbox"/> Priority</li> <li>III <input type="checkbox"/> Non-establishment of opinion with regard to novelty, inventive step and industrial applicability</li> <li>IV <input type="checkbox"/> Lack of unity of invention</li> <li>V <input checked="" type="checkbox"/> Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement</li> <li>VI <input type="checkbox"/> Certain documents cited</li> <li>VII <input type="checkbox"/> Certain defects in the international application</li> <li>VIII <input checked="" type="checkbox"/> Certain observations on the international application</li> </ul>

Date of submission of the demand  15/01/1999	Date of completion of this report  20.09.99
Name and mailing address of the international preliminary examining authority:   European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx. 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  Kolitz, R  Telephone No. +49 89 2399 8481



INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT

International application No. PCT/EP98/03583

I. Basis of the report

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

Description, pages:

1-28 as originally filed

Claims, No.:

1-19 as originally filed

2. The amendments have resulted in the cancellation of:

the description, pages:  
 the claims, Nos.:  
 the drawings, sheets:

3.  This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

1. Statement

Novelty (N)	Yes: Claims 1-19
	No: Claims
Inventive step (IS)	Yes: Claims 1-19
	No: Claims
Industrial applicability (IA)	Yes: Claims 1-19
	No: Claims

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP98/03583

**2. Citations and explanations**

**see separate sheet**

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

**see separate sheet**

**Section V:**

**Reasoned statement with regard to novelty, inventive step and industrial applicability Article 33 (1) to (4) PCT:**

D1 WO-A-9714749

D2 US-A-5415931 \*

D3 US-A-5242968 \*

\*) cited in the light of Rule 64.1a) PCT

1. The present invention relates to a composite plastics composition comprising a specific amount of a particulate crosslinked polymer of a defined particle size range being dispersed within a thermoplastic matrix, wherein the crosslinked polymer comprises a specific amount of inert filler and crosslinker and is visually differentialble from the thermoplastic matrix (claims 1-11 and 15) and a process for preparing it (claims 12-14) and an extruded sheet material and a thermoformed product made thereof (claims 18-19).
2. The present application meets the requirements of Article 33 (1) -(3) PCT because the subject-matter of claims 1-19 is novel and inventive vis-à-vis the prior art cited in the search report.

The problem of the present application may be regarded as to provide a further composite plastics composition having a mineral- like appearance which can be fabricated by conventional thermoplastic processing methods such as sheeet extrusion and injection molding.

No indication was given in the prior art that the combination of specifc amounts of a particulate crosslinked polymer of a defined particle size range being dispersed within a thermoplastic matrix could be used to solve this problem.

D1 discloses a particulate crosslinked polymer of a defined particle size range being dispersed within a thermoset matrix. D1 reports that different approches lead to too much swelling such that the viscosity becomes unworkable.

According to D2 and D3 a crosslinked particulate polymer is dispersed in a

monomer/ polymer syrup of MMA in PMMA. After a swelling process of the particles the mixture is cured and a particle-matrix interpenetrating network is formed which is not thermoplastic.

None of the above-mentioned documents teaches or suggests a composite plastics composition which combines a crosslinked particulate polymer with a thermoplastic matrix, a process for preparing it and an extruded sheet material and a thermoformed product made thereof having all the features of present claims 1-19.

Therefore the novelty and the presence of an inventive step may be acknowledged for the subject-matter of the present claims 1-19.

3. The present application meets the requirements of Article 33 (1) and (4) PCT because the subject-matter of claims 1-19 is also industrially applicable.

### **Section VIII:**

#### **Remarks concerning clarity , Article 6 PCT:**

The following clarity deficiencies must be overcome in the regional phase:

1. The claims must be clear in themselves since the claims (and not the description) define the matter for which protection is sought, Art.6 PCT.
  - 1.1. The definitions of the expressions "cross-linked polymer" and "inert filler" are unclear and overlap with each other. The cross-linked polymer (C) can be regarded as inert filler(F) and vice versa. These overlapping definitions render the scope of the claims unclear. Since (C) should comprise a specific amount of (F) it is absolutely necessary to distinguish between them and to define both expressions properly e.g . by incorporation of the subject-matter of claims 6 and 9 into claim 1.
  - 1.2. Moreover, the expression "cross-linker" is not clearly defined in claim 1. It is necessary to define what a cross-linker is e.g. by incorporation of the subject-matter of claim 8 into claim 1.
  - 1.3. The definition of an impact modifier is not clear in claim 2 and should be clarified by incorporation of the subject-matter of claim 4.
2. The expression "substantially" in connection with ranges e.g. in claims 1, 10, 11,

12, 13 and in the description renders the scope of the claims unclear and should therefore be deleted.

3. Claims 1, 11 and 15 do not meet the requirements of Article 6 PCT since they have been drafted as separate independent claims, however appear to relate effectively to the same subject-matter. The aforementioned claims therefore lack conciseness. Moreover, lack of clarity of the claims as a whole arises, since the plurality of independent claims makes it difficult to determine the matter for which protection is sought, and places an undue burden on others seeking to establish the extent of the protection.

In order to overcome this objection, it would appear appropriate to file an amended set of claims defining the relevant subject-matter in terms of a single independent claim in each category followed by dependent claims covering features which are merely optional.

4. All examples 3A to 3C in Table 2 on page 26 have an amount of 35% cross-linked polymer and 65% thermoplastic polymer. Although all of them appear to be examples of the invention example 3B, only, gives good results, see "sheet extrusion" and "sheet appearance" in Table 2, whereby the result of examples 3A and 3C does not solve the problem of the present application to provide a further composite composition having granite-like appearance. On the contrary examples 3A and 3C show cross-linked particles "melted into the matrix" or a "poor process" with "stranding and pelletizing problems" and the final composite exhibits "no granite texture" or "rough surface".

It is therefore unclear why Examples 3A and C are examples of the invention and why example 3B, only, gives good results.

- 4.1. The comments on page 28, last sentence, refer to examples 3D and 3G in Table 3. However, in Table 3 examples 3D and 3G do not exist.

## PATENT COOPERATION TREATY

PCT

REC'D 20 SEP 1999

WIPO

PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)

Applicant's or agent's file reference N.74722 MAM/lp	<b>FOR FURTHER ACTION</b>		See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)
International application No. PCT/EP98/03583	International filing date (day/month/year) 15/06/1998	Priority date (day/month/year) 19/06/1997	
International Patent Classification (IPC) or national classification and IPC C08L101/00			
Applicant ELF ATOCHEM S.A. et al.			

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.

2. This REPORT consists of a total of 6 sheets, including this cover sheet.

This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).

These annexes consist of a total of sheets.

3. This report contains indications relating to the following items:

- I  Basis of the report
- II  Priority
- III  Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV  Lack of unity of invention
- V  Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI  Certain documents cited
- VII  Certain defects in the international application
- VIII  Certain observations on the international application

Date of submission of the demand 15/01/1999	Date of completion of this report 20.09.99
Name and mailing address of the international preliminary examining authority: European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer Kolitz, R Telephone No. +49 89 2399 8481



**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP98/03583

**I. Basis of the report**

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

**Description, pages:**

1-28 as originally filed

**Claims, No.:**

1-19 as originally filed

2. The amendments have resulted in the cancellation of:

the description,      pages:  
 the claims,      Nos.:  
 the drawings,      sheets:

3.  This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes: Claims 1-19
	No: Claims
Inventive step (IS)	Yes: Claims 1-19
	No: Claims
Industrial applicability (IA)	Yes: Claims 1-19
	No: Claims

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/EP98/03583

**2. Citations and explanations**

**see separate sheet**

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

**see separate sheet**

**Section V:**

**Reasoned statement with regard to novelty, inventive step and industrial applicability Article 33 (1) to (4) PCT:**

D1 WO-A-9714749

D2 US-A-5415931 \*

D3 US-A-5242968 \*

\*) cited in the light of Rule 64.1a) PCT

1. The present invention relates to a composite plastics composition comprising a specific amount of a particulate crosslinked polymer of a defined particle size range being dispersed within a thermoplastic matrix, wherein the crosslinked polymer comprises a specific amount of inert filler and crosslinker and is visually differential from the thermoplastic matrix (claims 1-11 and 15) and a process for preparing it (claims 12-14) and an extruded sheet material and a thermoformed product made thereof (claims 18-19).
2. The present application meets the requirements of Article 33 (1) -(3) PCT because the subject-matter of claims 1-19 is novel and inventive vis-à-vis the prior art cited in the search report.

The problem of the present application may be regarded as to provide a further composite plastics composition having a mineral- like appearance which can be fabricated by conventional thermoplastic processing methods such as sheet extrusion and injection molding.

No indication was given in the prior art that the combination of specific amounts of a particulate crosslinked polymer of a defined particle size range being dispersed within a thermoplastic matrix could be used to solve this problem.

D1 discloses a particulate crosslinked polymer of a defined particle size range being dispersed within a thermoset matrix. D1 reports that different approaches lead to too much swelling such that the viscosity becomes unworkable.

According to D2 and D3 a crosslinked particulate polymer is dispersed in a

monomer/ polymer syrup of MMA in PMMA. After a swelling process of the particles the mixture is cured and a particle-matrix interpenetrating network is formed which is not thermoplastic.

None of the above-mentioned documents teaches or suggests a composite plastics composition which combines a crosslinked particulate polymer with a thermoplastic matrix, a process for preparing it and an extruded sheet material and a thermoformed product made thereof having all the features of present claims 1-19.

Therefore the novelty and the presence of an inventive step may be acknowledged for the subject-matter of the present claims 1-19.

3. The present application meets the requirements of Article 33 (1) and (4) PCT because the subject-matter of claims 1-19 is also industrially applicable.

### **Section VIII:**

#### **Remarks concerning clarity , Article 6 PCT:**

The following clarity deficiencies must be overcome in the regional phase:

1. The claims must be clear in themselves since the claims (and not the description) define the matter for which protection is sought, Art.6 PCT.
  - 1.1. The definitions of the expressions "cross-linked polymer" and "inert filler" are unclear and overlap with each other. The cross-linked polymer (C) can be regarded as inert filler(F) and vice versa. These overlapping definitions render the scope of the claims unclear. Since (C) should comprise a specific amount of (F) it is absolutely necessary to distinguish between them and to define both expressions properly e.g. by incorporation of the subject-matter of claims 6 and 9 into claim 1.
  - 1.2. Moreover, the expression "cross-linker" is not clearly defined in claim 1. It is necessary to define what a cross-linker is e.g. by incorporation of the subject-matter of claim 8 into claim 1.
  - 1.3. The definition of an impact modifier is not clear in claim 2 and should be clarified by incorporation of the subject-matter of claim 4.
2. The expression "substantially" in connection with ranges e.g. in claims 1, 10, 11,

12, 13 and in the description renders the scope of the claims unclear and should therefore be deleted.

3. Claims 1, 11 and 15 do not meet the requirements of Article 6 PCT since they have been drafted as separate independent claims, however appear to relate effectively to the same subject-matter. The aforementioned claims therefore lack conciseness. Moreover, lack of clarity of the claims as a whole arises, since the plurality of independent claims makes it difficult to determine the matter for which protection is sought, and places an undue burden on others seeking to establish the extent of the protection.

In order to overcome this objection, it would appear appropriate to file an amended set of claims defining the relevant subject-matter in terms of a single independent claim in each category followed by dependent claims covering features which are merely optional.

4. All examples 3A to 3C in Table 2 on page 26 have an amount of 35% cross-linked polymer and 65% thermoplastic polymer. Although all of them appear to be examples of the invention example 3B, only, gives good results, see "sheet extrusion" and "sheet appearance" in Table 2, whereby the result of examples 3A and 3C does not solve the problem of the present application to provide a further composite composition having granite-like appearance. On the contrary examples 3A and 3C show cross-linked particles "melted into the matrix" or a "poor process" with "stranding and pelletizing problems" and the final composite exhibits "no granite texture" or "rough surface".

It is therefore unclear why Examples 3A and C are examples of the invention and why example 3B, only, gives good results.

4.1. The comments on page 28, last sentence, refer to examples 3D and 3G in Table 3. However, in Table 3 examples 3D and 3G do not exist.



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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(71) Applicant ( <i>for all designated States except US</i> ): <b>ELF ATOCHEM S.A. [FR/FR]; 4 &amp; 8, cours Michelet, La Défense 10, F-92080 Puteaux (FR).</b>  (72) Inventors; and (75) Inventors/Applicants ( <i>for US only</i> ): <b>YANG, Shijun [CN/US]; 1803 Perrin Court, Maple Glen, PA 19002 (US). KEATING, Paul, Joseph [US/US]; 2727 Avenue "A", Newportville, PA 19056 (US).</b>  (74) Agents: <b>MARSHALL, Monica, Anne et al.; J.A. Kemp &amp; Co., 14 South Square, Gray's Inn, London WC1R 5LX (GB).</b>			<b>Published</b> <i>With international search report.</i> <i>Before the expiration of the time limit for amending the claims and to be republished in the event of the receipt of amendments.</i>

(54) Title: **PLASTIC COMPOSITION**

(57) Abstract

Composite plastics compositions based on the dispersion of selected cross-linked polymers in certain thermoplastic matrices are disclosed. The composite compositions are readily formed and processed by thermal extrusion processes versus conventional casting processes. Preferred composite plastics compositions based on cross-linked poly(alkyl (meth)acrylate) polymers and modified poly(alkyl (meth)acrylate) thermoplastics are especially useful in the preparation of synthetic architectural materials having a mineral-like appearance, such as that of granite.



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## PLASTIC COMPOSITION

This invention relates to composite plastics  
5 compositions useful in the preparation of simulated natural substances, such as stone-like and mineral-like materials. In particular the present invention involves the use of specific crosslinked polymers together with certain thermoplastic matrices to provide composite plastics  
10 compositions having a mineral-like appearance, especially an appearance similar to that of granite.

There currently exists a need for synthetic materials that mimic the appearance of mineral-like or ceramic-like materials, such as natural stone, and in particular 15 granite, for use in the manufacture of flooring, tiles, counter tops, sinks, spas, sanitaryware, architectural articles and other ornamental materials. For example, acrylic "granite" sheet products useful in spas, sanitaryware and outdoor applications are currently 20 prepared by cell or continuous casting processes where small granulates are suspended in monomer or monomer/polymer mixtures and then "cured," such as is described in U.S. Patent No. 5,304,592 and WO 97/14749. These thermoset materials can not be further fabricated by 25 conventional thermoplastic processing methods (such as sheet extrusion and injection molding operations); furthermore, the thermoset processes currently require special handling steps or raw material limitations to uniformly suspend the granulates in the polymerizing matrix. 30 during the "cure" step. Reuse and recycling of waste and off-grade materials, such as trimmings, from prior art sheet materials is impractical due to the inherent

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intractability of the thermoset materials. Prior art "granite" sheet products prepared by casting methods generally have poor impact strength, for example brittleness, and are limited in their adaptability to various formulation or processing methods for producing final articles. In addition, the prior art materials often provide poor adhesion to substrate surfaces resulting in subsequent "delamination" of composite articles.

It is, therefore, desirable to be able to provide thermoplastic materials that may be fabricated by conventional thermoplastic equipment (such as extrusion or injection molding) for use in spas and outdoor architectural applications; there is a need for an economical and efficient method for reuse and recycling of waste materials that is not available with current thermoset casting processes.

The problem addressed by the present invention is to overcome the deficiencies of prior methods used to prepare mineral-like plastics by providing materials that are processable by less labor intensive methods, such as coextrusion or extrusion, while also improving the economics of the processing by allowing for reuse and recycling of waste materials.

The present invention provides a composite plastics composition comprising a particulate crosslinked polymer dispersed within a thermoplastic matrix, wherein (a) the composite plastics composition comprises 10 to 45 weight percent of the crosslinked polymer, based on the weight of the composite plastics composition, and the crosslinked polymer has a particle size substantially from 0.2 to 1.2 millimeters; (b) the crosslinked polymer comprises 0.1 to 15 weight percent inert filler and 0.1 to 20 weight percent

crosslinker, based on the total weight of crosslinked polymer; and (c) the crosslinked polymer is visually differentiable from the thermoplastic matrix.

The present invention further provides a process for 5 preparing a composite plastics composition comprising (a) preparing a crosslinked polymer comprising 0.1 to 15 weight percent inert filler and 0.1 to 20 weight percent crosslinker, based on the weight of crosslinked polymer; (b) comminuting the crosslinked polymer to particles having 10 a particle size substantially from 0.2 to 1.2 millimeters; (c) dispersing 10 to 45 weight percent of the particles of crosslinked polymer within 55 to 90 weight percent of a thermoplastic matrix by a heat processing treatment; and (d) recovering the composite plastics composition as a 15 particulate material.

In another aspect, the present invention provides a composite plastics composition comprising a particulate crosslinked polymer dispersed within a thermoplastic matrix, wherein (a) the composite plastics composition comprises more than 20 and up to 40 weight percent of the crosslinked polymer, based on the weight of the composite plastics composition, and the particle size of the crosslinked polymer is substantially from 0.3 to 1.2 millimeters; (b) the crosslinked polymer comprises (i) from 20 95 to 99.5 weight percent (meth)acrylic monomer units selected from one or more of methyl methacrylate, ethyl acrylate and acrylic acid; (ii) from 0.5 to 5 weight percent crosslinker units selected from one or more of allyl methacrylate, ethylene glycol dimethacrylate and divinylbenzene; and (iii) from 0.3 to 5 weight percent 25 inert filler selected from one or more of titanium dioxide, iron oxide, alumina, carbon black, pigments and silica, 30

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based on the total weight of crosslinked polymer; (c) the thermoplastic matrix comprises (i) 50 to 60 weight percent poly(alkyl (meth)acrylate) comprising a copolymer of 80 to 99 weight percent methyl methacrylate monomer units and 1 to 20 weight percent ( $C_1-C_{10}$ ) alkyl acrylate monomer units, based on the weight of poly(alkyl (meth)acrylate); and (ii) 40 to 50 weight percent impact modifier comprising a multi-stage sequentially produced polymer, based on the weight of thermoplastic matrix; and (d) the crosslinked polymer is visually differentiable from the thermoplastic matrix.

In other aspects, the present invention provides a plastics composite composition prepared according to the process described above and an article of manufacture comprising an extruded sheet material resulting from extrusion of the composite plastics composition described above.

The process of the present invention is useful for preparing a range of composite plastics compositions suitable for use in forming simulated mineral-like articles. Common to each of the composite compositions is the dispersion of a particulate crosslinked polymer within a thermoplastic matrix. We have found that composite plastics compositions based on selected crosslinked polymers, having selected crosslinking levels and a selected particle size range, result in unexpectedly improved extrusion process performance and the ability to provide "granite-like" plastics articles as compared with prior art plastics using cell-casting processes.

As used herein, the term "thermoplastic" refers to polymers that are reversibly deformable (able to be softened) after being heated above their softening or glass transition temperatures and then cooled; these materials

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are capable of being repeatedly melt processed in plastic manufacturing machinery such as, for example, injection molding, extrusion, blow molding, compression molding and rotational molding. As is generally accepted by those skilled in the art, thermoplastic polymers include, for example, acrylonitrile/butadiene/styrene (ABS) terpolymer, acrylonitrile/styrene/acrylate (ASA) copolymer, polycarbonate, polyester, methyl methacrylate/butadiene/styrene (MBS) copolymer, high impact polystyrene (HIPS), acrylonitrile/acrylate copolymer, acrylonitrile/methyl methacrylate copolymer, impact modified polyolefins, impact modified polyvinyl chloride (PVC) and impact modified polymethacrylates.

As used herein, the term "thermoset" refers to polymers that are irreversibly deformable after they have been prepared in an initial configuration, that is, once the polymer is formed by chemical crosslinking (usually thermally induced) it is no longer amenable to thermal processing into other physical forms. As is generally accepted by those skilled in the art, thermoset polymers are polymers that include crosslinking as part of their preparation or have been subjected to crosslinking reactions as part of a post-treatment step, for example, allyl ester polymers, epoxy resins, crosslinked acrylic polymers and crosslinked styrenic polymers produced by suspension, emulsion, continuous-cast or cell-cast polymerization methods.

As used herein, the term "thermoforming" refers to the processing of polymers into 3-dimensional molded forms from flat plastic preformed materials, such as film or sheet, under the influence of heat, pressure or vacuum or combinations thereof.

As used herein, the term "particulate material" refers to any material in the form of separate particles or divided fragments, such as, for example, pellets, beads, powders, granules and chips.

As used herein, the term "alkyl (meth)acrylate" refers to either the corresponding acrylate or methacrylate ester; similarly, the term "(meth)acrylic" refers to either acrylic or methacrylic acid and the corresponding derivatives, such as esters or amides. As used herein, all percentages referred to will be expressed in weight percent (%), based on total weight of polymer or composition involved, unless specified otherwise. As used herein, the term "copolymer" or "copolymer material" refers to polymer compositions containing units of two or more monomers or monomer types. As used herein, "extrusion blended" and "extrusion compounded" are used synonymously and refer to the intimate mixing of two materials by melt extrusion. The composite plastics compositions of the present invention use selected crosslinked polymers in combination with selected thermoplastic matrix. Crosslinked polymers useful in the present invention include, for example, crosslinked vinyl polymers (prepared from monoethylenically unsaturated monomers and crosslinked condensation polymers (such as polyepoxy resins and polyesters, for example, poly(butylene terephthalate) and poly(ethylene terephthalate)). Thermoset polymers represent one class of crosslinked polymers useful in the present invention. Suitable monoethylenically unsaturated monomers useful in preparing particulate material useful in the present invention include vinylaromatic monomers, ethylene

and substituted ethylene monomers.

Suitable vinylaromatic monomers include, for example, styrene and substituted styrenes, such as  $\alpha$ -methylstyrene, vinyltoluene, ortho-, meta- and para-methylstyrene, ethylvinylbenzene, vinylnaphthalene and vinylxylenes. The vinylaromatic monomers can also include their substituted counterparts, for example, halogenated derivatives, that is, containing one or more halogen groups (such as fluorine, chlorine and bromine).

Another class of suitable monoethylenically unsaturated monomers is ethylene and substituted ethylene monomers, for example:  $\alpha$ -olefins such as propylene, isobutylene and long chain alkyl  $\alpha$ -olefins (such as (C<sub>10</sub>-C<sub>20</sub>)alkyl olefins); vinyl alcohol esters such as vinyl acetate and vinyl stearate; vinyl halides such as vinyl chloride, vinyl fluoride, vinyl bromide, vinylidene chloride, vinylidene fluoride and vinylidene bromide; vinyl nitriles such as acrylonitrile and methacrylonitrile; acrylic acid and methacrylic acid and derivatives such as corresponding amides and esters; maleic acid and derivatives such as corresponding anhydride, amides and esters; fumaric acid and derivatives such as corresponding amides and esters; itaconic and citraconic acids and derivatives such as corresponding anhydrides, amides and esters.

A preferred class of monomers useful in preparing the crosslinked polymers of the present invention are (meth)acrylic monomers, particularly (C<sub>1</sub>-C<sub>22</sub>)alkyl (meth)acrylate monomers. Examples of the alkyl (meth)acrylate monomer where the alkyl group contains from 1 to 6 carbon atoms are methyl methacrylate (MMA), methyl acrylate and ethyl acrylate (EA), propyl methacrylate,

butyl methacrylate (BMA), butyl acrylate (BA), isobutyl  
methacrylate (IBMA), hexyl and cyclohexyl methacrylate,  
cyclohexyl acrylate and combinations thereof.  
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Examples of alkyl (meth)acrylate monomers where the  
alkyl group contains from 7 to 22 carbon atoms are 2-  
ethylhexyl acrylate (EHA), 2-ethylhexyl methacrylate,  
(IDMA, based on branched (C10)alkyl isomer mixture),  
undecyl methacrylate, dodecyl methacrylate, isodecyl methacrylate,  
lauryl methacrylate (also known as myristyl methacrylate, tetradearyl  
methacrylate and combinations thereof. Also useful are: dodecyl-pentadecyl methacrylate, tetradearyl  
pentadecyl methacrylate, dodecyl methacrylate, tetradearyl  
tridecyl, tetradearyl and branched isomers of dodecyl,  
lauryl-myristyl methacrylate (LMA), a mixture of dodecyl,  
and tetradecyl methacrylates.  
Additional (C7-C22)alkyl (meth)acrylate monomers  
include hexadecyl methacrylate (also known as cetyl  
methacrylate), heptadecyl methacrylate, octadecyl  
methacrylate (also known as stearyl methacrylate), behenyl  
nonadecyl methacrylate, eicosyl methacrylate, behenyl  
methacrylate and combinations thereof; also useful are  
cetyl-eicosyl methacrylate (CEMA), a mixture of hexadecyl,  
octadecyl, and eicosyl methacrylate; and cetyl-stearyl  
methacrylate.  
Suitable crosslinking monomers include, for example,  
ethylene glycol dimethacrylate, polyethylene glycol  
diacrylate and dimethacrylate, propylene glycol  
dimethacrylate and diacrylate, glycidyl methacrylate,  
divinylbenzene, triallyl isocyanurate, N-

(hydroxymethyl)acrylamide, allyl acrylate, allyl methacrylate, N,N'-methylene diacrylamide and dimethacrylamide, triallyl citrate, trimethylolpropane triacrylate, trimethylolpropane trimethacrylate, and diethyleneglycol divinyl ether. Preferred crosslinkers are allyl methacrylate, ethylene glycol dimethacrylate and divinylbenzene. The amount of crosslinking monomer is generally from 0.1 to 20%, typically from 0.5 to 10%, preferably from 0.5 to 5%, more preferably from 1 to 4% and most preferably from more than 1.5% up to 3%, based on the total weight of the crosslinked polymer, that is, combined weight of monoethylenically unsaturated monomer and the crosslinking monomer.

Suitable free-radical initiators useful in the present invention are any of the well known free-radical-producing compounds such as peroxy and hydroperoxy initiators, including, for example, acetyl peroxide, benzoyl peroxide, lauroyl peroxide, caproyl peroxide, cumene hydroperoxide, 1,1-di(tert-butylperoxy)-3,3,5-trimethylcyclohexane, tert-butyl peroxyisobutyrate, tert-butyl peracetate, tert-butyl peroxy pivalate (TBPV) and tert-butyl peroctoate (TBP, also known as tert-butylperoxy-2-ethylhexanoate). Also useful, for example, are azo initiators such as azodiisobutyronitrile (AIBN), azodiisobutyramide, 2,2'-azo-bis(2,4-dimethylvaleronitrile), azo-bis( $\alpha$ -methylbutyronitrile) and dimethyl-, diethyl- or dibutyl azo-bis(methylvalerate). The initiator concentration is typically between 0.01 and 5%, preferably from 0.05 to 2% and more preferably from 0.1 to 1%, by weight based on the total weight of the monomers. In addition to the initiator, one or more promoters may also be used; preferably the promoters are hydrocarbon-soluble.

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Alternatively, little or no free-radical initiator may be used and the polymerization is then thermally induced by heating the monomer mixture.

The crosslinked polymer also contains inert filler  
5 that may be conveniently added to a cell casting syrup  
before starting the polymerization. Suitable inert  
fillers, include, for example, alumina (including hydrated  
forms), titanium dioxide, zinc oxide, zinc sulfide, iron  
oxide, barium sulfate, zirconium silicate, strontium  
10 sulfate, calcium carbonate, carbon black, powdered glass,  
silica, clay and talc. Preferred inert fillers include  
titanium dioxide, iron oxide, carbon black, silica,  
alumina, pigments and combinations thereof; most preferred  
is titanium dioxide. Conventional pigments or colorants  
15 include organic dyes (for example azo, anthraquinone,  
perinone, quinoline, pyrazolone, dioxazine, isoindoline,  
phthalocyanine, quinacridone and coumarin derivatives) and  
inorganic salts (cadmium, chromates, iron blue, cobalt blue  
and ultramarine blue). Typically, the amount of inert  
20 filler is from 0.1 to 15%, preferably from 0.2 to 10% and  
more preferably from 0.3 to 5%, based on the total weight  
of crosslinked polymer. The purpose of the inert filler is  
to enhance the visual differentiation between the  
crosslinked polymer component and the thermoplastic matrix  
25 component to provide the "mineral-like" or "granite-like"  
appearance of the finished composite plastics composition.

Optionally, the crosslinked polymer may contain  
conventional adjuvants, known to those skilled in the art,  
for various purposes, for example: dyes, pigments,  
30 antioxidants, ultraviolet stabilizers, dispersants,  
processing aids (such as spray drying aids, lubricants and  
mold-release agents), flame retardants, polymerization

rate-moderators and viscosity controlling materials. In addition, these same conventional adjuvants may be conveniently added at later stages in the production of the composite plastics composition, for example, during preparation of the thermoplastic material used as the thermoplastic matrix (described below) or during the production of the composite plastics composition itself where the crosslinked polymer and thermoplastic material are extrusion blended or compounded.

Typically, the particulate crosslinked polymers used according to the present invention are selected from the group consisting of crosslinked poly(alkyl (meth)acrylate), crosslinked poly(vinylaromatic), crosslinked polyester, crosslinked polyolefin, mixtures and corresponding copolymers thereof. Preferred crosslinked polymers are crosslinked poly(alkyl (meth)acrylates) and crosslinked polystyrene where the crosslinked polymer comprises 90 to 99.5% monomer units selected from one or more of vinylaromatic monomer and (meth)acrylic monomer and 0.5 to 10% crosslinker, based on the weight of crosslinked polymer.

The crosslinked polymer material is conveniently prepared by a cell casting process, for example. In a typical cell casting process a monomer syrup containing the monoethylenically unsaturated monomers, a crosslinking agent and a free-radical initiator is subjected to a temperature suitable for polymerization, depending on the monomers and initiators used. Preferred monoethylenically unsaturated monomers are alkyl (meth)acrylate monomers, for example, (C<sub>1</sub>-C<sub>4</sub>)alkyl (meth)acrylates, such as MMA (typically 80 to 99.5% and preferably from 95 to 99.5%, based on the total weight of monomers), methylacrylate or

EA (typically from zero to 20%, preferably from 1 to 8% and more preferably from 0.5 to 5%, based on the total weight of monomers), propyl methacrylate, BMA, BA, IBMA and combinations thereof. Optionally, acrylic or methacrylic acid may be included, typically from zero to 2%, based on the total weight of monomers.

After the crosslinked polymer is prepared, such as by cell cast polymerization, it is comminuted to a particulate material by known processes, for example, chipping, crushing, grinding, shredding or any granulation method; if suspension or emulsion polymerization processes are used to prepare the crosslinked polymer, conventional isolation processes used to recover the crosslinked polymer in particulate form include, for example, filtration, coagulation and spray drying. The dimensions of the particulate crosslinked polymer are typically from 0.2 to about 1.2 millimeters (mm) or about 16 to 70 U.S. standard mesh. Generally, the particle size of the particulate crosslinked polymer is substantially from 0.25 to 1.2 mm (16 to 60 mesh), more preferably from 0.3 to 1.0 mm (16 to 50 mesh), and most preferably from 0.4 to 0.85 mm (20 to 40 mesh).

When the particle size of the crosslinked polymer is smaller than about 70 mesh or larger than about 16 mesh, the extrusion blending of the crosslinked polymer is "stranding" problems, and the desired "speckle-like" or "mineral-like" effect may not be readily attained in the thermoplastic matrix becomes problematic, for example, final decorative or architectural article.

Thermoplastic materials useful to provide the thermoplastic matrix of the present invention include, for example, alkyl (meth)acrylate polymers and copolymers.

having little or no crosslinker. The thermoplastic materials are conveniently prepared by conventional cell casting or melt extrusion processes and are typically provided in particulate form. In addition, the 5 thermoplastic materials may be prepared by conventional bulk (for example, continuous flow stirred tank reactor (CFSTR) processes), solution, suspension or emulsion polymerization techniques, in which case conventional isolation processes used to recover the polymer in 10 particulate form include, for example filtration, coagulation and spray drying. Conditions for polymerization of monomers to produce the thermoplastic material are similar to those described above for cell casting polymerization, except that little or no 15 crosslinker is involved; when melt extrusion methods are used, the thermoplastic material is conveniently isolated in a particulate form, such as pellets or granules. Suitable alkyl (meth)acrylate monomers include, for example, the (C<sub>1</sub>-C<sub>22</sub>)alkyl (meth)acrylates monomers 20 described above for the use in preparing the crosslinked polymers. Preferably the thermoplastic matrix material comprises a polymer or copolymer of methyl methacrylate (MMA); typical copolymers include 80 to 99% MMA and 1 to 20%, preferably 1 to 5%, of (C<sub>1</sub>-C<sub>10</sub>)alkyl acrylates, such as 25 methyl acrylate and ethyl acrylate (EA). A suitable commercially available poly(methyl methacrylate) type thermoplastic matrix material is Plexiglas V-grade molding powder, such as Plexiglas VO-825, VO-825HID, VO-45, VO-52 and VO-920.

30 Additional suitable thermoplastic polymers include, for example, ABS terpolymer, ASA copolymer, polycarbonate, polyester (such as poly(butylene terephthalate) and

poly(ethylene terephthalate)), MBS copolymer, HIPS, acrylonitrile/acrylate copolymer, acrylonitrile/methyl methacrylate copolymer, impact modified polyolefins and impact modified PVC. More preferably the thermoplastic matrix material is an impact modified polymethacrylate.

The thermoplastic matrix may be based entirely on the aforementioned thermoplastic polymers or the thermoplastic matrix may optionally contain modifier additives, such as impact modifiers, in addition to the aforementioned thermoplastic polymers. In general, the thermoplastic matrix comprises 50 to 100% poly(alkyl (meth)acrylate) and zero to 50% impact modifier, based on the weight of thermoplastic matrix. Typically, the thermoplastic matrix contains 25 to 100%, preferably 30 to 70%, more preferably 45 to 60% and most preferably 50 to 60%, thermoplastic polymer, such as poly(alkyl (meth)acrylate); and zero to 75%, preferably 30 to 70%, more preferably 40 to 55% and most preferably 40 to 50%, impact modifier, based on the total weight of thermoplastic matrix. Suitable impact modifiers include, for example, elastomeric polymers such as graft polymers of methyl methacrylate and styrene on butadiene (MBS), graft polymers of acrylonitrile and styrene on butadiene (ABS), copolymers of styrene and butadiene, poly(butyl acrylate) and poly(2-ethylhexyl acrylate) and copolymers thereof, copolymers of butyl acrylate and methyl acrylate, terpolymers of butyl acrylate/styrene/methyl methacrylate, chlorinated polyethylene, acrylate block polymers, styrene block polymers, ethylene/propylene/diene copolymer (EPDM), ethylene/vinyl acetate copolymers, acrylonitrile/styrene/acrylic ester terpolymers, styrene-maleic anhydride copolymers and core-shell multi-stage

sequentially-produced polymers. Preferred impact modifiers include MBS polymers, core-shell multi-stage sequentially-produced polymers, and styrene and acrylate block polymers.

Typical core-shell multi-stage polymers useful as impact modifiers for thermoplastic polymers include, for example, those disclosed in U.S. Patent No. 3,793,402. The multi-stage sequentially-produced polymers are characterized by having at least three stages in a sequence of a non-elastomeric first stage, an elastomeric second stage and a non-elastomeric third stage. Preferably the first stage polymer has a glass transition temperature ( $T_g$ ) greater than about 25°C, preferably greater than about 60°C, and is a polymer comprising (a) 70 to 100%, preferably 85 to 99.9%, monomer units of one or more of (C<sub>1</sub>-C<sub>4</sub>)alkyl (meth)acrylates (preferably MMA and EA), styrene, substituted styrene, acrylonitrile and methacrylonitrile, based on the weight of the first stage polymer; (b) zero to 10%, preferably 0.05 to 5% and more preferably 0.5 to 2%, monomer units of a copolymerizable polyfunctional crosslinking monomer; (c) zero to 10%, preferably 0.05 to 5%, monomer units of a copolymerizable graftlinking monomer, such as the allyl, methallyl and crotyl esters of monoethylenically unsaturated monocarboxylic and dicarboxylic acids, for example allyl methacrylate; and (d) zero to 30%, preferably 0.1 to 15%, of other copolymerizable monoethylenically unsaturated monomers.

The second stage polymer is prepared in the presence of the first stage polymer and preferably has a  $T_g$  of less than about 25°C, preferably less than about 10°C, if it were to be prepared alone, that is in the absence of the presence of the first stage polymer. Typically the second

stage polymer comprises (a) 50 to 99.9%, preferably 70 to 99.5%, monomer units of one or more of (C<sub>1</sub>-C<sub>8</sub>)alkyl (meth)acrylates, (preferably (C<sub>1</sub>-C<sub>4</sub>)alkyl acrylates, particularly BA), butadiene and substituted butadienes (such as isoprene, chloroprene and 2,3-dimethylbutadiene), based on the weight of the second stage polymer; (b) zero to 49.9%, preferably 0.5 to 30%, monomer units of a copolymerizable monoethylenically unsaturated monomer, such as styrene and substituted styrene; (c) zero to 5% monomer units of a copolymerizable polyfunctional crosslinking monomer, such as ethylene glycol diacrylate and divinylbenzene; and (d) 0.05 to 5% monomer units of a copolymerizable graftlinking monomer, such as those described above.

The third stage polymer is prepared in the presence of the product of the first and second stage polymers and preferably has a T<sub>g</sub> greater than about 25°C, preferably greater than about 50°C, if it were to be prepared alone, that is in the absence of the presence of the product of the first and second stage polymers. Typically the third stage polymer comprises (a) 70 to 100%, preferably 85 to 99.9%, monomer units of one or more of (C<sub>1</sub>-C<sub>4</sub>)alkyl (meth)acrylates, styrene, substituted styrene, acrylonitrile and methacrylonitrile, based on weight of the second stage polymer; (b) zero to 30, preferably 0.1 to 15%, monomer units of a copolymerizable monoethylenically unsaturated monomer; (c) zero to 10%, preferably 0.05 to 5%, monomer units of a copolymerizable polyfunctional crosslinking monomer; and (d) zero to 5% monomer units of a copolymerizable graftlinking monomer, such as those described above.

Typically the multi-stage polymer comprises at least

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three stages in a sequence of 10 to 40%, preferably 20 to 40%, of the first stage; 20 to 60%, preferably 30 to 50%, of the second stage; and 10 to 70%, preferably 20 to 50%, of the third stage, based on the total weight of the multi-stage polymer.

Other copolymerizable mononethylenically unsaturated monomers referred to above include alkyl (meth)acrylates, alkoxy (meth)acrylates, hydroxyalkyl (meth)acrylates, cyanoethyl (meth)acrylates, (meth)acrylamides, (meth)acrylic acids and vinyl aromatics, for example.

Typical copolymerizable polyfunctional crosslinking monomers include, for example, ethylene glycol dimethacrylate and diacrylate, 1,3-butylene glycol dimethacrylate and diacrylate, 1,4-butylene glycol dimethacrylate and diacrylate, propylene glycol dimethacrylate, divinylbenzene, trimethylolpropane triacrylate, trimethylolpropane trimethacrylate and diethyleneglycol divinyl ether. Preferred crosslinkers are ethylene glycol dimethacrylate and divinylbenzene. For the purpose of the preparing the multi-stage sequentially-produced polymers, the crosslinkers are selected from polyfunctional monomers where the crosslinking groups have similar reactivities, such as those just described.

Polyfunctional "crosslinking" monomers where the functional groups have different reactivities, such as allyl methacrylate and methallyl and crotyl esters of monoethylenically unsaturated monocarboxylic and dicarboxylic acids, are referred to as "graftlinking" monomers when used in preparation of the multi-stage sequentially-produced polymers described herein; however they are considered to be among the general group of "crosslinking" monomers useful in the preparation of other

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crosslinked polymers, such as those used to provide the particulate crosslinked polymers of the present invention.

Preferably, the multi-stage polymer is an emulsion polymer comprising monomer units of methyl methacrylate in the first stage, monomer units selected from one or more of butadiene, styrene and (C<sub>1</sub>-C<sub>8</sub>)alkyl acrylates in the second stage, and monomer units selected from one or more of (C<sub>1</sub>-C<sub>4</sub>)alkyl methacrylates, styrene and acrylonitrile in the third stage.

The multi-stage polymers may be prepared by a number of well-known techniques, for example, by emulsion polymerization, where a subsequent stage monomer mixture is polymerized in the presence of a previously formed product. For example, "sequentially produced" or "sequentially polymerized" refer to polymers prepared in aqueous dispersion or emulsion form where successive monomer charges are polymerized onto or in the presence of a preformed latex prepared by the polymerization of a prior monomer charge and stage.

The blend of the multi-stage polymer (as an impact modifier) with the thermoplastic polymer can be accomplished by any known method, such as dispersing the multi-stage polymer in a monomer mixture used to prepare the thermoplastic polymer or in a monomer-polymer syrup mixture which together would provide the desired thermoplastic polymer. Alternatively, the multi-stage polymer can be placed in a casting mix in the form of an emulsion, suspension or dispersion in water or in an organic carrier; the water or organic carrier can then be removed before or after casting into the final thermoplastic polymer form. The multi-stage polymer may also be blended with the thermoplastic polymer by extrusion

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compounding. Additional specific methods and details of blending the thermoplastic polymer and impact modifiers are disclosed in U.S. Patent No. 3,793,402.

A preferred thermoplastic matrix material is impact modified poly(methyl methacrylate) commercially available as Plexiglas DR101, MI-5 and MI-7 molding powder.

Plexiglas (North and South America, Oroglas in Europe and Asia) is a trademark of Rohm and Haas Company, Philadelphia, PA, USA.

In one embodiment of the present invention, the composite plastics compositions of the present invention are prepared by compounding or blending the crosslinked polymer with the thermoplastic material by dispersing particles of crosslinked polymer within a matrix of thermoplastic material using a suitable heat processing treatment. Suitable heat processing treatments include, for example, extrusion blending, hot-melt kneading and hot-melt batch mixing. For example, the crosslinked polymer particles may be melt processed by hot melt extrusion blending or compounding with thermoplastic particles and the resultant composite plastic composition is recovered in particulate form, such as pellets. Hot-melt batch mixing may include dispersion of the crosslinked particles into a melt of the thermoplastic matrix in a conventional batch mode, such as a stirred kettle; alternatively, the thermoplastic may be heated and mixed with a carrier solvent, such as toluene, and then batch mixed with the crosslinked particles, followed by flash evaporation of the solvent; the resultant composite plastic composition can then be granulated by conventional means. The resultant pellets are typically translucent to clear granules containing distinct "speckle-like" particles, the latter

being representative of the crosslinked polymer used in the compounding process.

Typically 10 to 45%, preferably 15 to 40% and more preferably greater than 20 to 40%, crosslinked polymer particles are combined with 55 to 90%, preferably from 60 to 85% and more preferably from 60 to less than 80%, thermoplastic particles, based on the combined weight of crosslinked polymer and thermoplastic particles. The extrusion blending process allows for a relatively uniform distribution of the thermoset material throughout the thermoplastic matrix without requiring similar densities of the two materials, as is typically required in conventional casting processes used in the prior art. Preferably a low-shear screw design is used to minimize processing problems, such as residual monomer odor, vent plugging, flowability problems and stranding problems.

Extrusion temperatures are typically in the range of 220° to 260°C and the extruder screw design should provide low shear to prevent the loss of the distinct granite-like appearance and to maintain particle integrity. The cooling bath temperature (strand takeup) is typically maintained at about 60° to 70°C and the vent section of the extruder may be vacuumed to minimize pelletization problems.

During the extrusion blending process, control of the particle size of crosslinked polymer is important for optimum processing and ultimate "granite-like" appearance of the finished articles. When the particles are very large, that is, greater than about 1.2 mm or 16 mesh, "stranding" problems may develop during sheet extrusion or extrusion blending processes. When the particles are much smaller than about 0.2 mm or 70 mesh, the extruder die may plug frequently and stranding problems may develop; in

addition, poor "granite-like" appearance occurs.

If the crosslinking level is too low, that is, below about 0.5%, the crosslinked polymer particles may "smear" into thermoplastic matrix material after multiple passes during extrusion, resulting in blurred or non-differentiated mineral-like appearance. If the crosslinking level is too high, that is, above about 10%, the yield of desired particle size during the comminution step is reduced due to generation of fine particles smaller than about 0.2 mm (70 mesh) and the sheet surfaces of the final article may be rough and require a secondary treatment such as press polishing. Preferably, the crosslinker level is from 0.5 to 5% and more preferably from 1 to 4%.

An example of the importance of the degree of crosslinking in the particulate material regarding the maintenance of particle integrity during processing can be seen in the multiple extrusion processes typically involved in preparing monolithic and multilayed composite sheets.

When prior art particles ("C" particles from Safas Corp., mixture of thermoplastic and thermoset material - see Example 3E-3G) were extrusion blended with thermoplastic matrix material at a low concentration (less than 10%), the final sheet provided a hazy speckle-like appearance (not "granite-like"). At a 25% use rate, about half of the "C" particles were smeared into thermoplastic matrix resulting in a non-differentiated final product appearance. When the use rate of "C" particles was above 30%, extrusion process problems were observed (power surges, poor flow, stranding problem, extruder pluggage) as well as complete loss of granite-like effect in the composite plastic material. Composite plastic compositions prepared using the "C"

particles discussed above (thermoset/thermoplastic particles disclosed in U.S. Patent No 5,304,592) undergo loss of granite-like appearance under the extrusion processing conditions described in Example 3, apparently due to insufficient particle integrity.

Simulated mineral articles are prepared from the composite plastics compositions of the present invention by heat treatment into a physical form selected from the group consisting of sheet, laminated sheet and molded material.

Suitable heat treatment processes include, for example, melt extrusion, coextrusion, blow molding, sheet forming and thermoforming.

The composite plastics compositions of the present invention can be extruded into monolithic sheet for indoor or outdoor applications, or coextruded with other high-impact grade thermoplastics such as ABS terpolymer, ASA copolymer, polycarbonate, MBS copolymer, HIPS, acrylonitrile/acrylate copolymer, acrylonitrile/methyl methacrylate copolymer, impact modified polyolefins and impact modified PVC, to produce a multilayer composite sheet useful for spa, sanitary ware, countertops, bathroom and kitchen fixtures, wall decorations and other thermoforming applications. The composite plastics compositions may also be injection molded into different forms for other applications such as facets, frames, door handles, window frames, sinks, shower stalls, building panels, plumbing fixtures, tiles, refrigerator walls, floor coverings and decorative moldings.

The invention is further illustrated by way of example in the following Examples. All ratios, parts and percentages are expressed by weight unless otherwise specified, and all reagents used are of good commercial

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quality unless otherwise specified. Abbreviations used in the Examples and Tables are listed below with the corresponding descriptions.

5	MMA	=	Methyl Methacrylate
	EA	=	Ethyl Acrylate
	AA	=	Acrylic Acid
	ALMA	=	Allyl Methacrylate
10	AIBN	=	Azodiisobutyronitrile
	TBP	=	tert-Butyl Peroctoate
	TBPV	=	tert-Butyl Peroxypivalate
	TiO <sub>2</sub>	=	Titanium Dioxide

**Example 1** Preparation of Crosslinked Polymer

15 A monomer mixture was prepared by combining crosslinker monomer (ALMA), alkyl (meth)acrylate monomer (MMA), (meth)acrylic comonomer (EA, AA), inert filler (TiO<sub>2</sub>, silica, carbon black), free-radical initiator (AIBN, TBP, TBPV mixture, 0.05%) polymerization rate-moderator (terpinolene, 0.01%) and lubricant (2% stearic acid). All 20 % values are by weight based on total weight of monomers.

25 The ingredients listed above (monomer mixture) were mixed in a glass casting cell or polyvinyl alcohol (PVA) casting bag and then subjected to a controlled temperature of 60°C for 14 hours, followed by a post-cure (heating at 130°C for 2 hours). The finished crosslinked polymer was recovered by disassembly of the cell or removal of the PVA bag and was then granulated into particulate form using any conventional grinding method, for example, crushing, 30 attrition mill or Cumberland cutter. The granulated crosslinked polymer was then screened to a desired particle size distribution.

Table I summarizes crosslinked polymers (component % based on weight of crosslinked polymer, monomers plus

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crosslinker) evaluated in the composite plastic compositions of the present invention. Crosslinked polymers typically contained less than 1% residual monomer and were screened after comminution to particulate form, with a typical isolated yield of 65 to 80% particles of 0.4 mm to 1mm (-18+40 U.S. standard mesh).

Table I

## Compositions of Crosslinked Polymers

ID	Monomers <sup>a</sup>	Crosslinker <sup>b</sup>	Inert Filler <sup>c</sup>
1A	94.5/3.1/2.1	0.3	3
1B	93.0/3.0/2.0	2.0	3
1C	93.0/3.1/0.0	3.9	3
1D	95.0/3.1/0.0	1.9	2/2 <sup>d</sup>
1E <sup>e</sup>	96/4/0	0.0	2.6

**a** = MMA/EA/AA

**b** = ALMA

**c** = Titanium Dioxide

**d** = Silica/Carbon Black

**e** = Uncrosslinked, weight average molecular weight approx  $3 \times 10^6$

**Example 2** Preparation of Thermoplastic Matrix Material

The thermoplastic matrix material used in evaluating the composite plastic compositions of the present invention was impact modified poly(methyl methacrylate) commercially available as Plexiglas DR101 molding powder from Rohm and Haas Co, Philadelphia, PA, USA.

**Example 3** Preparation of Composite Plastic Composition

The particulate crosslinked polymer (0.4 to 1 mm granules) of Example 1 were extrusion blended (using a single- or twin-screw extruder at 220 to 260°C) with the

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thermoplastic material (pellets of approximately 3 to 6 mm in diameter and length) of Example 2 (in the relative amounts indicated) into the composite plastic composition of the present invention (see Table 2). The pelletized composite plastic compositions were then sheet extruded and evaluated for their aesthetic and processing characteristics. Examples 3A-3D represent the present invention and 3E-3G represent comparative examples that replace the particulate crosslinked polymer of the present invention with thermoplastic/thermoset particles based on U.S. Patent No. 5,304,592 (provided by Safas Corp.).

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Table 2

ID	Crosslinked Polymer	Thermoplastic Material	Sheet Extrusion	Sheet Appearance
3A	Ex 1A (35%)	Ex 2 (65%)	Crosslinked particles melted into matrix	No granite texture
3B	Ex 1B (35%)	Ex 2 (65%)	Good processing	Granite texture and smooth surface
3C	Ex 1C (35%)	Ex 2 (65%)	Poor process, stranding and pelletizing problems	Rough surface
3F **	"C" Particles <sup>a</sup> (25%)	Ex 2 (75%)	Majority of particles melted into matrix	Poor granite texture and poor particle/matrix differentiation*
3G **	"C" Particles <sup>a</sup> (32%)	Ex 2 (68%)	Poor process, stranding and pelletizing problems	No granite texture, opaque*
3H	Ex 1B/1D (< 10%)	Ex 2 (>90%)	Good processing	Transparent, little or no granite texture
3J	Ex 1B/1D (>45%)	Ex 2 (<55%)	Poor process, stranding and pelletizing problems	Rough surface
3K	Ex 1E (25-35%)	Ex 2 (65-75%)	Good processing	No granite texture

\* = appearance from Carver press "button"

15        \*\* = comparative composite plastic composition, not of the present invention

a = thermoplastic/thermoset particles from Safas Corp., -25+35 ANN

**Example 4** Physical Properties of Composite Plastics Compositions

Composite plastics compositions based on different crosslinked polymers (prepared according to Example 1) and the thermoplastic matrix material of Example 2 were fabricated into test samples of sheet (injection molding and sheet extrusion) or film (Carver press) and evaluated for overall appearance and impact strength.

The Falling Dart [dart: 1.36 kilogram (3-pound), 0.63 centimeter, cm (0.25 inch, in) radius; sample of 15.2 cm (6 in) x 15.2 cm (6 in) x 0.32 cm (0.125 in)] and Notched Izod (23°C, 73°F) tests were conducted according to ASTM method D256 (published by the American Society for Testing and Materials) as measures of impact strength.

Table 3

Composite Plastics Composition	Falling Dart Impact Test Total energy, joules (foot-pounds)	Notched Izod joule/centimeter (foot-pound/inch)
Ex 1B/1D (15%)/Ex 2 (85%)	10.4 (7.7)	0.42 (0.79)
Ex 1B/1D (30-37%)/Ex 2 (63-70%)	3.9 (2.9)	0.29-0.30 (0.54-0.56)

In addition to the sheet extrusion process used to evaluate the "granite-like" effect, the appearance of the compounded composite material was also visually examined using a "button" prepared on a Carver press according to the following conditions: 65 grams of composite plastic

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composition pellets were placed in an "X" configuration in a 12.7 cm x 12.7 cm x 0.25 cm thick (5 in x 5 in x 0.1 in thick) aluminum mold between chrome-plated plates; the press was subjected to 1 minute preheat at 188°C (370°F) under  $3.45 \times 10^6$  pascals (Pa) pressure (500 pounds per square inch/psig), 1 minute heat at 188°C (370°F) under  $8.27 \times 10^7$  Pa (12,000 psig) and 1 minute cool at 10°C (50°F) under  $8.27 \times 10^7$  Pa (12,000 psig). The appearances of the Carver press "buttons" for composite plastic compositions of the present invention and that of the prior art were consistent with those described in Table 3 for Examples 3D and 3G, respectively.

**CLAIMS**

1. A composite plastics composition comprising a particulate crosslinked polymer dispersed within a thermoplastic matrix, wherein:

5 (a) the composite plastics composition comprises 10 to 45 weight percent of the crosslinked polymer, based on the weight of the composite plastics composition, and the crosslinked polymer has a particle size substantially from 0.2 to 1.2 millimeters;

10 (b) the crosslinked polymer comprises 0.1 to 15 weight percent inert filler and 0.1 to 20 weight percent crosslinker, based on the total weight of crosslinked polymer; and

15 (c) the crosslinked polymer is visually differentiable from the thermoplastic matrix.

2. A composite plastics composition according to claim 1 wherein the thermoplastic matrix comprises 50 to 100 weight percent poly(alkyl (meth)acrylate) and zero to 50 weight percent impact modifier, based on the weight of thermoplastic matrix.

25 3. A composite plastics composition according to claim 2 wherein the poly(alkyl (meth)acrylate) comprises a copolymer of 80 to 99 weight percent methyl methacrylate monomer units and 1 to 20 weight percent (C<sub>1</sub>-C<sub>10</sub>)alkyl acrylate monomer units, based on total weight of the poly(alkyl (meth)acrylate).

30 4. A composite plastics composition according to claim 2 or 3 wherein the impact modifier is a multi-stage

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sequentially-produced polymer comprising at least three stages in a sequence of a non-elastomeric first stage, an elastomeric second stage and a non-elastomeric third stage.

5 5. A composite plastics composition according to claim 4 wherein the multi-stage polymer is an emulsion polymer comprising monomer units of methyl methacrylate in the first stage, monomer units selected from one or more of butadiene, styrene and (C<sub>1</sub>-C<sub>8</sub>)alkyl acrylates in the second 10 stage, and monomer units selected from one or more of (C<sub>1</sub>-C<sub>4</sub>)alkyl methacrylates, styrene and acrylonitrile in the third stage.

15 6. A composite plastics composition according to any one of the preceding claims wherein the crosslinked polymer comprises 90 to 99.5 weight percent monomer units selected from one or more of vinylaromatic monomer and (meth)acrylic monomer and 0.5 to 10 weight percent crosslinker, based on the weight of crosslinked polymer.

20 7. A composite plastics composition according to claim 6 wherein the (meth)acrylic monomer is selected from one or more of methyl methacrylate, methyl acrylate, ethyl acrylate, acrylic acid and butyl methacrylate.

25 8. A composite plastics composition according to any one of the preceding claims wherein the crosslinker is selected from one or more of allyl methacrylate, ethylene glycol dimethacrylate and divinylbenzene.

30 9. A composite plastics composition according to any one of the preceding claims wherein the inert filler is

selected from one or more of titanium dioxide, iron oxide, alumina, pigments, carbon black and silica.

10. A composite plastics composition according to any one  
5 of the preceding claims wherein the particle size of the  
crosslinked polymer is substantially from 0.3 to 1.2  
millimeters.

11. A composite plastics composition comprising a  
10 particulate crosslinked polymer dispersed within a  
thermoplastic matrix, wherein:

15 (a) the composite plastics composition comprises more  
than 20 and up to 40 weight percent of the crosslinked  
polymer, based on the weight of the composite plastics  
composition, and the particle size of the crosslinked  
polymer is substantially from 0.3 to 1.2 millimeters;

20 (b) the crosslinked polymer comprises (i) from 95 to  
99.5 weight percent (meth)acrylic monomer units  
selected from one or more of methyl methacrylate,  
ethyl acrylate and acrylic acid; (ii) from 0.5 to 5  
weight percent crosslinker units selected from one or  
more of allyl methacrylate, ethylene glycol  
dimethacrylate and divinylbenzene; and (iii) from 0.3  
25 to 5 weight percent inert filler selected from one or  
more of titanium dioxide, iron oxide, alumina, carbon  
black and silica, based on the total weight of  
crosslinked polymer;

30 (c) the thermoplastic matrix comprises (i) 50 to 60  
weight percent poly(alkyl (meth)acrylate) comprising a  
copolymer of 80 to 99 weight percent methyl  
methacrylate monomer units and 1 to 20 weight percent

(C<sub>1</sub>-C<sub>10</sub>)alkyl acrylate monomer units, based on the weight of poly(alkyl (meth)acrylate); and (ii) 40 to 50 weight percent impact modifier comprising a multi-stage sequentially produced polymer, based on the weight of thermoplastic matrix; and

5 (d) the crosslinked polymer is visually differentiable from the thermoplastic matrix.

12. A process for preparing a composite plastics  
10 composition comprising:

(a) preparing a crosslinked polymer comprising 0.1 to 15 weight percent inert filler and 0.1 to 20 weight percent crosslinker, based on the weight of crosslinked polymer;

15 (b) comminuting the crosslinked polymer to particles having a particle size substantially from 0.2 to 1.2 millimeters;

(c) dispersing 10 to 45 weight percent of the particles of crosslinked polymer within 55 to 90 weight percent of a thermoplastic matrix by a heat  
20 processing treatment; and

(d) recovering the composite plastics composition as a particulate material.

25 13. A process according to claim 12 wherein the particles of step (b) have a particle size substantially from 0.3 to 1.2 millimeters.

14. A process according to claim 12 or 13 wherein the heat  
30 processing treatment of step (c) is selected from one or more of extrusion blending, hot-melt kneading and hot-melt batch mixing.

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15. A composite plastics composition prepared by the process claimed in any one of claims 12 to 14.

16. A process for preparing a simulated mineral article comprising forming, with heat treatment of a composite plastics composition as claimed in any one of claims 1 to 11 into a sheet, laminated sheet or molded material.

17. A process according to claim 16 wherein the heat treatment is selected from the group consisting of melt extrusion, coextrusion, blow molding, sheet forming and thermoforming.

18. An extruded sheet material resulting from extrusion of a composite plastics composition as claimed in any one of claims 1 to 11.

19. A thermoformed product of a composite plastics composition as claimed in any one of claims 1 to 11.

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# INTERNATIONAL SEARCH REPORT

International Application No

PCT/EP 98/03583

A. CLASSIFICATION OF SUBJECT MATTER  
 IPC 6 C08L101/00 B29C47/10 // (C08L101/00, 101:00)

According to International Patent Classification(IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C08L B29C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	WO 97 14749 A (ICI ACRYLICS INC) 24 April 1997 cited in the application see page 9, line 5 - line 25 -----	1

Further documents are listed in the continuation of box C.

Patent family members are listed in annex.

### Special categories of cited documents :

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Information on patent family members

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WO 9714749 A	24-04-1997	AU 7432696 A EP 0856032 A	07-05-1997 05-08-1998



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